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RADIOACTIVITY OF ROCKS

ROBLEY D. EVANS AND CLARK GOODMAN

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ABSTRACT

Although numerous previous measurements of the radioactivity of terrestrial materials have been made, most of these researches are of qualitative value only in their applications to geology, geophysics, and cosmology because of inadequate recog-

nition of the analytical care necessary in order to obtain reliable results. In the present study of the radioactivity of terrestrial materials, a systematic program of standardization, calibration, and interchecking has been followed throughout. As part of an international intercalibration among the various workers in this field and a general program of helium age research, several hundred radioactivity measurements have been made. These results represent the most reliable collection of radioactivity determinations which have yet been made within the range of concentrations involved. By combining these newer measurements with the limited number of well-authenticated earlier analyses available, average values have been obtained as follows: $1.37 \pm 0.17 \times 10^{-12}$ g Ra/g for 43 acidic igneous rocks, $0.51 \pm 0.05 \times 10^{-12}$ g Ra/g for 7 intermediate igneous rocks, $0.38 \pm 0.03 \times 10^{-12}$ g Ra/g for 54 basic igneous rocks, and $0.57 \pm 0.08 \times 10^{-12}$ g Ra/g for 28 sedimentary rocks; $3.0 \pm 0.3 \times 10^{-6}$ g U/g, $13 \pm 2.0 \times 10^{-6}$ g Th/g, and a Th/U ratio of 5.0 for 26 acidic igneous rocks; $1.4 \pm 0.2 \times 10^{-6}$ g U/g, $4.4 \pm 1.2 \times 10^{-6}$ g Th/g, and a Th/U ratio of 2.6 for 6 intermediate igneous rocks; $0.96 \pm 0.11 \times 10^{-6}$ g U/g, $3.9 \pm 0.6 \times 10^{-6}$ g Th/g, and a Th/U ratio of 4.0 for 34 basic igneous rocks. These values are substantially lower than those obtained by Jeffreys in a compilation of most of the measurements reported prior to 1936. The present averages show a more marked decrease of radioactivity with increasing basicity, the Th/U ratios are considerably greater than those compiled by Jeffreys and are in better agreement with those to be expected from geochemical considerations. Two ultrabasic rocks were found to have radioactivities comparable to the low values for iron meteorites. Specific inaccuracies in earlier investigations have been discovered. Estimates are made of the rate of production of heat by radioactive decay based on the above average values for the different rock types.

INTRODUCTION

Knowledge of the radioactive content of terrestrial materials is of fundamental importance in many phases of geology, geophysics, and cosmology. The problem of the internal heat of the earth, and its relation to the source of magmatic energy, involves accurate information concerning the rate at which radioactive heat is being generated in the various types of rocks. The variation of radioactivity with rock type is also of interest in the study of the differentiation of magmas. The distribution of the radioactive elements among different minerals within a given rock may have important implications in geochemistry. Geological age measurements by radioactive methods depend upon accurate determination of the content of the long-lived parent radioactive elements in rocks and minerals. The radioactivity of sea water and marine sediments is illuminating in certain aspects of oceanography. The increasing applications of radioactive methods in geophysical prospecting focus attention on the need for further information on the radioactivity of rocks, particularly those of sedimentary origin. Measurements of the radioactivity of crude oils and associated sediments suggest that the chemical effects of alpha radiation may be important in the genesis of petroleum.

While it is true that the determination of relative magnitudes of radioactivity has some value, the truly significant interpretations in geology, and in other fields of science, will be possible only after the radioactivities of terrestrial materials have been established on an absolute basis

and have been evaluated in terms of the accurately known physical constants of the radioactive elements. Toward this goal, several hundred radioactivity measurements have been made on a wide variety of terrestrial specimens. Precautions have been taken to eliminate as many of the usual uncertainties as possible. A systematic program of standardization, calibration, and interchecking has been followed throughout this research. As the result of this care, it can be said with considerable confidence that these data represent the most reliable collection of radioactivity measurements which have yet been made within the range of concentrations involved.

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RADIOACTIVITY MEASUREMENTS OF ROCKS

GENERAL CONSIDERATIONS

Most of the radioactivity measurements of rocks reported in this paper were made as part of the general program of helium age research. Other determinations resulted from investigations on the genesis of petroleum, the standardization of radioactivity measurements, and the international intercalibration program. Practicality dictated that each measurement have many-fold applications if possible. Consequently, the rock specimens show a somewhat less systematic geologic distribution than would have been selected had these measurements been made solely for the purpose of establishing the averages indicated for the different lithologic types.

RADIUM MEASUREMENTS

Techniques.—The direct-fusion method of releasing radon from rocks as developed by Evans (1935, p. 102) has been employed, with essentially no modification, in all the radium measurements. Two techniques were used in determining the ionization effects of the radon and its immediate decay products. Total ionization measurements were made with the

double ionization-chamber, string-electrometer, photographic recording instrument (Evans, 1935, p. 104). Other determinations employed a more recently developed apparatus that counts the individual alpha rays, emitted in a single cylindrical ionization-chamber, by the use of the usual type of vacuum-tube electrometer (Penick, 1935, p. 115). Photo-

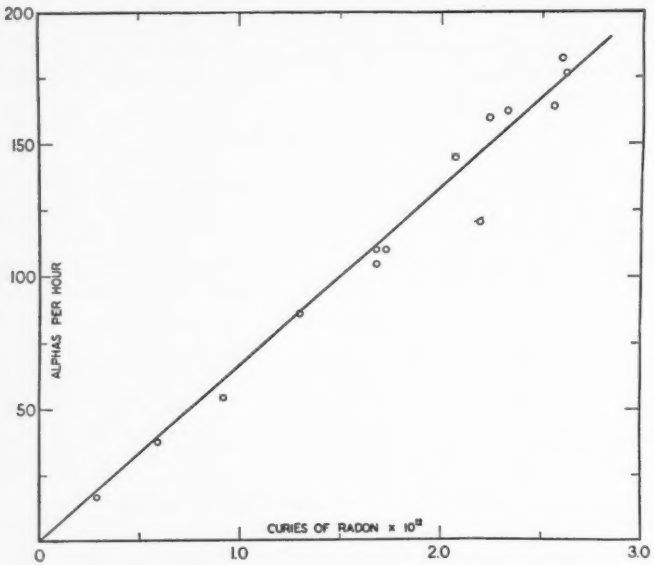

FIGURE 1.—*Calibration of alpha-counting radon apparatus*

graphic recording affords continuous observation up to 20 hours on a single sheet of bromide paper 6 by 37 inches. This apparatus is a liberally modified version of the one described by Poole (1938, p. 595).

Calibration.—The calibration of the double ionization chamber apparatus has been discussed in detail elsewhere (Evans *et al.*, 1939, p. 937). The alpha-counting radium apparatus has also been calibrated with standard solutions prepared from the Lind-Evans radium standard. The variation in observed alpha-count with radon content is linear over the lower range of values found in rocks (Fig. 1). On a basis of 5 grams of sample, the quantity normally used, the upper end of the graph corresponds to a radium content of about 0.6×10^{-12} g Ra/g of rock. With increase in radium content above this value, the reliability of the alpha-count method decreases rapidly due to the lack of resolution

of the individual counts when using a vacuum-tube electrometer and ballistic galvanometer. For this reason the alpha-count apparatus has a narrower useful range than the ionization apparatus, but this limitation is compensated by the somewhat greater accuracy of the former for low radium values. A sufficient number of check measurements have been made in the intermediate region of activities to assure good agreement between the two apparatuses.

Measurements.—The observed radium contents of a number of rocks, expressed in the customary units of 10^{-12} g Ra/g, are arranged according to general lithologic types in five tables (Tables 1 to 5). Tables 1, 2, and 3 contain the results on acidic, intermediate, and basic rocks; Tables 4 and 5, metamorphic and sedimentary rocks. Two samples, 30B and 35B included in Table 3, are of particular interest not only in reflecting the unusually low radium content of ultrabasic rocks, but they also serve to indicate that the analytical techniques do not include appreciable contamination effects. Brief geologic notes for each rock specimen are given in a later section.

Accuracy.—An estimate of the over-all analytical probable error of an individual radium measurement can be obtained from a consideration of repeat measurements on uniform samples and on standard solutions used for calibration purposes. Using standard radium solutions for reference purposes, values of the calibration constant, K_E, obtained over a 2-year period, show a maximum spread in distribution of only 2.3 per cent from the mean (Evans *et al.*, 1939, Table II, p. 937). These observations indicate: (1) the degree of reliability of the ionization measurements and (2) that the electrical properties of the detection apparatus have not changed significantly during the measurements. Duplicate analyses of homogeneous rock samples afford a check on: (1) the reliability of the direct-fusion method of releasing radon and (2) the efficacy of the transfer of radon from the furnace to the ionization chamber for measurement. The results for multiple analyses of rock samples 3A in Table 1, 5B, 18B, 19B, and 28B in Table 3, and 9S in Table 5 are in agreement within 8 per cent of the mean in each case. Since these measurements were made on different portions of the same powdered sample of each rock, the interpretation given is that the parent element, uranium, and hence also its daughter element, radium, is fairly uniformly distributed throughout the hand specimens. After a rock is ground, different 5 to 10 gram portions appear to contain equal radium contents within the experimental uncertainty of measurement, which is estimated to be 8 per cent. At very low radium contents, the statistical fluctuations in the

TABLE 1.—*Radium measurements of acidic igneous rocks*

Sample No.	Rock specimen	Ra content in 10^{-12} g/g rock	
		Individual measurements	Mean value
1A	Brifosse granite	8.0 ±0.40	8.0 ±1.6
2A	Kumlinge granite	*3.4 ±0.2	3.8 ±0.5
		*4.1 ±0.2	
3A	Missourian granite	*3.17 ±0.03	3.1 ±0.1
		*3.14 ±0.05	
		*2.92 ±0.03	
		3.18 ±0.03	
$4A_1$	Chelmsford granite	4.50 ±0.05	2.5 ±0.1
$4A_2$	Chelmsford granite	*2.48 ±0.05	
		*2.04 ±0.03	
		*3.10 ±0.04	
		*2.40 ±0.03	
		*2.02 ±0.03	
5A	Rapakiwi granite	2.42 ±0.05	2.3 ±0.2
		*2.20 ±0.04	
6A	Granite	2.3 ±0.10	2.3 ±0.5
7A	Rockport granite	2.14 ±0.07	2.3 ±0.3
		1.96 ±0.06	
		2.81 ±0.10	
8A	Rockport xenolith	1.80 ±0.06	1.80 ±0.36
9A	Murray granite	1.34 ±0.03	1.34 ±0.27
$10A_1$	Creighton granite	1.64 ±0.04	1.25 ±0.25
$10A_2$	Creighton granite	1.20 ±0.05	
$10A_3$	Creighton granite	0.95 ±0.05	
		0.80 ±0.03	
11A	Granite, Franklin, Me.	1.24 ±0.05	1.24 ±0.25
12A	Quincy granite	0.94 ±0.03	0.94 ±0.19
13A	Granite	0.88 ±0.04	0.88 ±0.18
14A	Ava granite	*0.75 ±0.05	0.88 ±0.12
		*1.0 ±0.05	
15A	Fitchburg granite	0.88 ±0.03	0.88 ±0.18
16A	Granite, Kawatana, H. S.	0.87 ±0.01	0.87 ±0.18
17A	Kingston granite	0.41 ±0.04	0.36 ±0.05
		0.31 ±0.03	
18A	Azi granite	0.21 ±0.006	0.21 ±0.04
19A	Milford granite	*0.20 ±0.01	0.205±0.02
		*0.21 ±0.01	
20A	Ounasjok granite	*0.19 ±0.04	0.20 ±0.03
		*0.20 ±0.03	
21A	Granite	0.12 ±0.01	0.12 ±0.02
	Average for 21 granites:		1.69 ±0.20
22A	Rhyolite du Rosskopf	1.80 ±0.07	1.80 ±0.36
23A	Rhyolite	1.62 ±0.06	1.3 ±0.4
		0.93 ±0.03	
24A	Golden Gate rhyolite	1.23 ±0.05	1.23 ±0.25
25A	Porphyritic rhyolite	0.88 ±0.04	0.88 ±0.18
26A	Creighton aplite phase	3.13 ±0.07	3.13 ±0.63
27A	Aso pitchstone	1.47 ±0.01	1.47 ±0.30
28A	Obsidian Cliff	1.36 ±0.04	1.36 ±0.27
29A	Sudbury micropegmatite	1.13 ±0.02	1.01 ±0.11
		0.88 ±0.01	
	Average for 29 acidic rocks:		1.64 ±0.20

* Sample stored for 30 days or longer before radon was removed for measurement.

emission of alpha-particles dictate the precision of the measurements (Evans and Neher, 1934, p. 144).

Because of the indicated reliability for moderate radium values (0.1×10^{-12} g/g or above), variations between duplicate analyses in excess of 10 per cent are attributed to an inhomogeneous distribution

TABLE 2.—*Radium measurements of igneous rocks intermediate between acidic and basic*

Sample No.	Rock specimen	Ra content in 10^{-12} g/g rock	
		Individual measurements	Mean value
1I	Crescent Hill trachyte	0.90 ±0.03	0.90 ±0.18
2I	Bunsen Peak dacite	0.60 ±0.02	0.60 ±0.12
3I	Gray tuff	0.52 ±0.04	0.57 ±0.04
		0.53 ±0.04	
		0.66 ±0.05	
4I	La Motte granodiorite	0.57 ±0.04	0.57 ±0.11
5I	Tholeiite, Cleveland dike	0.44 ±0.03	0.44 ±0.09
6I	Hornblende granodiorite	0.24 ±0.04	0.29 ±0.04
		0.33 ±0.05	
7I	Quartz dolerite, Whin Sill	0.17 ±0.01	0.17 ±0.04
	Average value of 7 intermediates:		0.51 ±0.05

of radium, and hence of its parent element, uranium, in the sample. Such inhomogeneity appears to be usual rather than exceptional for rocks and introduces a sampling probable error which may greatly exceed the analytical uncertainty. In the tabulation of data, the analytical probable error, based on the residuals of hourly observations, is given after each individual measurement. The probable error of the mean of a number of individual measurements includes an observed, or estimated, field sampling uncertainty. In all single measurements, a sampling probable error of 20 per cent has been arbitrarily assigned when the observed value is listed as the mean for the particular rock body. For the few rock bodies that have been studied sufficiently to allow an estimate of their inhomogeneity, 20 per cent appears to represent the median variation in radioactive content.

The probable errors of the average radioactivities indicated for the various rock types were calculated by the usual statistical methods from the residuals of the mean values. It is understood that average values, obtained by repeating the measurements on the same number of rock specimens selected at random, will have a 50 per cent chance of lying within the range set by the probable error of the present average values. In attempting to estimate the average radioactivity of the earth's crust, much larger probable errors must be assigned to the various rock types, since such a small portion of the crust has been sampled and measured.

TABLE 3.—*Radium measurements of basic igneous rocks*

Sample No.	Rock specimen	Ra content in 10^{-12} g/g rock	
		Individual measurements	Mean value
1B	Porphyritic basalt	0.82 ±0.03	0.82 ±0.16
2B	Gravenoire basalt	0.64 ±0.04	0.64 ±0.13
3B	Crescent Hill basalt	0.39 ±0.01	0.39 ±0.08
4B	Giant's Causeway	0.39 ±0.03	0.39 ±0.08
5B	Columbia River basalt	*0.325±0.009	0.31 ±0.02
		*0.303±0.012	
		0.305±0.007	
		0.30 ±0.03	
6B	Oldwick basalt	0.25 ±0.01	0.21 ±0.04
		0.16 ±0.02	
7B	Geode Creek basalt	0.20 ±0.02	0.20 ±0.04
8B	Deccan trap	*0.218±0.006	0.19 ±0.03
		*0.15 ±0.01	
		0.20 ±0.01	
$9B_1$	Watchung basalt	0.18 ±0.01	0.17 ±0.03
$9B_2$	Watchung basalt	0.16 ±0.01	
10B	Overhanging Cliff	0.17 ±0.02	0.17 ±0.03
11B	Hawaiian basalt	0.18 ±0.02	0.15 ±0.03
		*0.11 ±0.01	
12B	Basalt	0.14 ±0.02	0.14 ±0.03
13B	Oxbow Creek basalt	0.13 ±0.02	0.13 ±0.03
	Average value for 13 basalts:		0.30 ±0.04
14B	Diabase (Horne Mine)	0.46 ±0.03	0.46 ±0.09
$15B_1$	Sudbury olivine diabase	0.39 ±0.04	0.34 ±0.08
$15B_2$	Sudbury olivine diabase	0.54 ±0.03	
		0.21 ±0.02	
$15B_3$	Sudbury olivine diabase	0.26 ±0.01	
$16B_1$	Palisade diabase	0.24 ±0.01	0.19 ±0.02
$16B_2$	Palisade diabase	0.19 ±0.01	
$16B_3$	Palisade diabase	0.19 ±0.01	
$16B_4$	Palisade diabase	0.15 ±0.01	
$16B_5$	Palisade diabase	0.17 ±0.01	
17B	Somerset County diabase	0.21 ±0.01	0.19 ±0.02
		*0.17 ±0.01	
18B	Virginia Triassic diabase	*0.17 ±0.03	0.18 ±0.01
		*0.17 ±0.005	
		0.186±0.004	
	Average value for 5 diabase rocks:		0.27 ±0.03
19B	Worthington norite	*0.85 ±0.04	0.76 ±0.11
		*0.70 ±0.04	
		*0.83 ±0.05	
		0.65 ±0.04	
		0.76 ±0.05	
$20B_1$	Sudbury norite	0.55 ±0.02	0.55 ±0.07
$20B_2$	Sudbury norite	0.54 ±0.02	
21B	Sudbury norite	0.25 ±0.02	0.25 ±0.05
22B	Stillwater norite	0.14 ±0.02	0.13 ±0.01
		0.11 ±0.03	
	Average value for 4 norites:		0.42 ±0.08

TABLE 3.—*Radium measurements of basic igneous rocks*—Continued

Sample No.	Rock specimen	Ra content in 10^{-12} g/g rock	
		Individual measurements	Mean value
$23B_1$	Sudbury trap	0.92 ±0.06	0.87 ±0.15
$23B_2$	Sudbury trap	0.82 ±0.05	
$24B_1$	Long Lake trap	0.47 ±0.04	0.47 ±0.08
$24B_2$	Long Lake trap	0.46 ±0.04	
25B	Beartooth trap	0.20 ±0.04	0.21 ±0.03
		0.21 ±0.01	
26B	Riasg Buidhe monchiquite	0.68 ±0.03	0.68 ±0.14
27B	Kilchattan monchiquite	0.32 ±0.02	0.32 ±0.06
28B	Blueberry Mt. gabbro-diorite	*0.168±0.005	0.16 ±0.01
		*0.15 ±0.007	
		0.17 ±0.01	
29B	Sudbury gabbro	0.096±0.006	0.096±0.02
30B	Dunite, Webster, N. C	*0.011±0.003	0.012±0.004
		*0.008±0.002	
		0.013±0.004	
		*0.010±0.002	
		*0.016±0.003	
31B	Kimberlite, S. Africa	*0.62 ±0.01	0.54 ±0.06
		*0.54 ±0.01	
		*0.48 ±0.02	
		0.54 ±0.01	
		*0.53 ±0.01	
32B	Dalhousie andesite	0.37 ±0.03	0.37 ±0.07
33B	Andesite	0.20 ±0.01	0.20 ±0.04
34B	Camptonite, Burlington, Vt	0.085±0.008	0.077±0.018
		0.104±0.01	
		0.041±0.007	
35B	Wutchumna diorite	0.000±0.005	0.00 ±0.01
	Average value for 35 basic rocks:		0.31 ±0.03

* Sample was stored for 30 days or longer before radon was removed for measurement.

TABLE 4.—*Radium measurements of metamorphic rocks*

Sample No.	Rock specimen	Ra content in 10^{-12} g/g rock	
		Individual measurements	Mean value
1M	Taroko-kyo gneiss	0.77 ±0.02	0.77 ±0.15
2M	Augen gneis	0.71 ±0.03	0.71 ±0.15
3M	Saastal eclogite	0.077±0.007	0.07 ±0.01
		0.072±0.006	

THORIUM MEASUREMENTS

Techniques.—In the past, the thorium content of terrestrial materials has usually been determined from thoron measurements (for example: Evans, 1935, p. 110; Urry, 1936, p. 34). However, measurement of the total radioactive content from observations of the rate of alpha-particle

TABLE 5.—*Radium measurements of sedimentary rocks*

Sample No.	Rock specimen	Ra content in 10^{-12} g/g rock	
		Individual measurements	Mean value
1S	Silurian bituminous limestone	*2.46 ±0.09	2.3 ±0.3
		*2.20 ±0.10	
$2S_1$	Shaly limestone	1.30 ±0.07	1.0 ±0.2
$2S_2$	Shaly limestone	0.78 ±0.06	
$3S_1$	Shaly limestone and sandstone	0.31 ±0.03	0.30 ±0.04
$3S_2$	Shaly limestone and sandstone	0.29 ±0.03	
4S	Dolomite	0.29 ±0.03	0.29 ±0.06
5S	Carthage limestone	*0.13 ±0.01	0.11 ±0.01
		*0.21 ±0.02	
		*0.08 ±0.01	
		0.12 ±0.01	
		*0.11 ±0.01	
		0.08 ±0.01	
		*0.08 ±0.01	
		*0.08 ±0.01	
		*0.13 ±0.01	
	Average value for 5 limestones:		0.80 ±0.20
$6S_1$	Sandstone	1.42 ±0.10	1.13 ±0.20
$6S_2$	Sandstone	1.17 ±0.06	
$6S_3$	Sandstone	0.80 ±0.05	
$7S_1$	Sandstone	0.82 ±0.06	0.63 ±0.10
$7S_2$	Sandstone	0.58 ±0.05	
$7S_3$	Sandstone	0.51 ±0.05	
8S	Shaly sandstone	0.67 ±0.05	0.67 ±0.15
9S	Berea sandstone	*0.24 ±0.01	0.23 ±0.01
		*0.23 ±0.01	
		*0.205±0.005	
		*0.23 ±0.008	
		*0.21 ±0.005	
$10S_1$	Sandstone	0.19 ±0.05	0.21 ±0.03
$10S_2$	Sandstone	0.22 ±0.02	
	Average value for 5 sandstones:		0.73 ±0.12
11S	Quartzite	*0.067±0.002	0.07 ±0.01
		*0.047±0.003	
		*0.068±0.004	
		0.084±0.005	
$12S_1$	Clay shale	1.15 ±0.07	1.09 ±0.09
$12S_2$	Clay shale	1.09 ±0.06	
$12S_3$	Clay shale	1.05 ±0.05	
$13S_1$	Shale	1.09 ±0.15	1.08 ±0.12
$13S_2$	Shale	1.07 ±0.10	
	Average value for 13 sedimentary rocks:		0.70 ±0.12

* Sample was stored for 30 days or longer before radon was removed for measurement.

emission affords a simplified method of determining the thorium content of rocks, when the radium content is known. The equation relating these quantities is simply:

$$Th = (\text{Activity Index}—1.04\ Ra)/0.0886 \quad (1)$$

where *Activity Index* = the number of alpha particles (helium nuclei) emitted per hour per milligram of rock.

Th = thorium content in 10^{-6} g/g of rock.

Ra = radium content in 10^{-12} g/g of rock.

The derivation of Equation (1) and a detailed description of the procedure used in the measurement of the activity index are included in the paper by Goodman and Evans (1941, p. 507).

In the application of Equation (1), it is tacitly assumed that radioactive equilibrium exists. Since 1 million years suffices to establish equilibrium in the uranium series, and a shorter time is sufficient for the actino-uranium and thorium series, from a time standpoint this assumption is applicable to all but very young rocks. Recent alteration and weathering may preferentially remove certain series members and upset equilibrium, but this effect appears rather unlikely for nearly all the specimens measured, particularly the igneous rocks.

Calibration.—The alpha activity measurements are not dependent upon radioactive standards. (Goodman and Evans, 1941, p. 507). Hence, frequent recalibration, as is routine in radium measurements, is not customary in this technique. Some internal standardization measurements have been made (Finney and Evans, 1935, p. 511) using rock powders containing added known amounts of chemically analyzed radioactive ores. The results of these observations are in good agreement with the theoretical predictions for the alpha-counting method. However, the most convincing evidence of reliability is the concurrence of activity index measurements based on separate radium and thorium analyses of powdered rock samples also measured for alpha activity (Evans *et al.*, 1939, Table VI, p. 939). A number of similar measurements have subsequently been made on the Palisade diabase, the results of which are included in Table 8.

These intercheck thorium analyses are based on measurements of thoron by the streaming method [1] and depend upon calibration with standard thorium solutions. No official thorium standards have ever been established.[2] In the past, the customary procedure has been to prepare reference standards from chemically analyzed thorium minerals, usually a monazite, thorianite, or thorite. As a temporary standard, the authors have adopted a solution prepared from an analyzed thorian-

[1] The method used is similar to that described by Urry (1936, p. 35) but with a number of changes in the detection apparatus and improvements in the chemical techniques.

[2] The National Research Council Committee on Standards of Radioactivity is endeavoring to fill this need. Through the co-operation of Drs. A. F. Kovarik and J. P. Marble, work is now in progress on the preparation of a primary thorium standard, which it is hoped will later be adopted as the primary international thorium standard.

ite furnished by the late Professor Herman Schlundt. This solution is considered to contain 4.18×10^{-6} g Th/cc and 2.1×10^{-6} g U/cc, with 0.7 per cent HNO_3 for protective purposes. Comparison with thorium standards prepared from other analyzed thorium minerals (Evans *et al.*, 1939, p. 939) indicate agreement within the experimental uncertainty of a few per cent.

Measurements.—In Tables 6, 7, and 8 are listed the individual measurements and resulting mean values for activity index determinations of a number of rock specimens,[3] most of which also have been analyzed for radium as reported in Tables 1, 2, and 3. The uranium content of a given specimen is calculated from the mean radium value using the conversion: $U = 2.84 \times 10^6$ Ra. The calculated thorium content results from substitution of the mean activity index and mean radium value in Equation (1). In the last column of these tables, the ratios of thorium to uranium have been listed. The Th/U ratio is an index of the relative activity of the two series in the production of helium, radiogenic lead, and heat in rocks as discussed in a later section of this paper and by Goodman and Evans (1941, p. 506).

Accuracy.—The remarks, concerning inhomogeneity, field sampling uncertainty, and calculation of probable errors, included in the discussion of radium measurements, are equally applicable to thorium measurements. In addition, the small amount of sample used (less than 100 milligrams) in an individual alpha activity determination increases the analytical uncertainty to about 20 per cent for most samples. In order to decrease this uncertainty to the point where it is comparable to that of the radium analyses, several alpha-count measurements are usually made on each sample.

DISCUSSION OF RESULTS

General statement.—In considering the radioactivity measurements of rocks presented in Tables 1 to 8, the reader may justly ask: Do these data merely add to the several hundred other results on similar materials obtained by Strutt, Joly, Poole, Piggot, Fletcher, Urry, Evans, and the numerous other investigators of the past 30 years, and hence merely define our knowledge of terrestrial radioactivity a little more sharply? The answer to this inquiry is that the present measurements have considerable more significance in that they constitute the most complete set of well-authenticated results which are now available. An ever increasing mass of evidence indicates that few of the earlier measurements are sufficiently reliable to allow other than the broadest of geological deductions. Whereas individual determinations have been reported as having

[3] All the activity indices were determined from alpha-counting measurements, except for some separate radium and thorium measurements made on samples 16B and 18B as reported in Table 8.

TABLE 6.—*Radioactivity measurements of acidic ig*

Sample No.	Rock specimen	Activity index in alphas/hr Individual measurements	Mean va
8A	Rockport xenolith	4.05 ±0.09	3.9 ±
		3.84 ±0.08	
30A	Mt. Manitou granite	3.2 ±0.1	3.2 ±
$10A_2$	Creighton granite	2.9 ±0.3	2.8 ±
		3.0 ±0.1	
$10A_3$	Creighton granite	2.5 ±0.2	
6A	Granite, Glen Cove, Colorado	2.80 ±0.06	2.8 ±
		2.70 ±0.07	
15A	Fitchburg granite	2.1 ±0.1	2.8 ±
		3.8 ±0.2	
		3.0 ±0.2	
		2.3 ±0.1	
31A	Lebanon granite	2.90 ±0.05	2.3 ±
		2.80 ±0.06	
		1.66 ±0.05	
		2.11 ±0.06	
		2.20 ±0.06	
8A	Rockport granite	2.18 ±0.08	2.1 ±
		2.07 ±0.06	
32A	Granite	2.0 ±0.1	2.0 ±
$12A_2$	Quincy granite	1.85 ±0.10	1.7 ±
		1.95 ±0.10	
$12A_1$	Quincy granite	1.42 ±0.08	
		1.52 ±0.07	
11A	Granite, Franklin, Maine	1.27 ±0.03	1.24 ±
		1.20 ±0.02	
21A	Granite, Winnipeg, Manitoba	0.37 ±0.04	0.36 ±
		0.35 ±0.04	
	Average for granites:		2.3 ±
	Number of samples:		(1
23A	Rhyolite	3.6 ±0.1	3.6 ±
24A	Golden Gate rhyolite	3.5 ±0.1	3.5 ±
28A	Obsidian Cliff	3.3 ±0.1	3.3 ±
33A	Pegmatite, Ruggles Mine	0.35 ±0.03	0.50 ±
		0.48 ±0.03	
		0.56 ±0.04	
		0.59 ±0.04	
	Average for acidics:		2.4 ±
	Number of samples:		(1

* Radium measurements by Evans and Raitt (1935, p. 172), who have also previously reported radiu

urements of acidic igneous rocks

ivity index in alphas/hr•mg		U in 10^{-6}g/g rock	Th in 10^{-6}g/g rock	Th/U ratio
ividual urements	Mean value			
5 ±0.09	3.9 ±0.5	6.5 ±0.9	17. ±4.	2.6
4 ±0.08				
±0.1	3.2 ±0.4	*3.8 ±0.5	20. ±4.	5.4
±0.3	2.8 ±0.3	3.5 ±0.7	17. ±4.	4.8
±0.1				
±0.2				
0 ±0.06	2.8 ±0.3	6.5 ±1.4	4.3 ±1.0	0.7
0 ±0.07				
±0.1	2.8 ±0.3	2.5 ±0.5	21. ±5.	8.4
±0.2				
±0.2				
±0.1				
0 ±0.05	2.3 ±0.3	.....	.....	...
0 ±0.06				
6 ±0.05				
1 ±0.06				
0 ±0.06				
8 ±0.08	2.1 ±0.3	5.1 ±1.0	2.6 ±0.5	0.5
7 ±0.06				
±0.1	2.0 ±0.4	.....	.....	...
5 ±0.10	1.7 ±0.3	2.7 ±0.5	8.1 ±2.0	3.0
5 ±0.10				
2 ±0.08				
2 ±0.07				
7 ±0.03	1.24 ±0.16	3.5 ±0.7	0.0 ±0.5	0.0
0 ±0.02				
7 ±0.04	0.36 ±0.07	0.34 ±0.06	2.7 ±0.8	7.8
5 ±0.04				
.....	2.3 ±0.2	3.8 ±0.4	10.3 ±1.7	3.7
.....	(11)	(9)	(9)	(9)
±0.1	3.6 ±0.8	4.6 ±0.9	22. ±5.	4.7
±0.1	3.5 ±0.8	3.5 ±0.7	25. ±5.	7.2
±0.1	3.3 ±0.7	3.9 ±0.8	21. ±5.	5.4
5 ±0.03	0.50 ±0.05	.....	.....	...
8 ±0.03				
6 ±0.04				
0 ±0.04				
.....	2.4 ±0.2	3.9 ±0.3	13.4 ±1.6	4.2
.....	(15)	(12)	(12)	(12)

reviously reported radium measurements on 6A and 21A.

urements of acidic igneous rocks

ivity index in alphas/hr•mg		U in 10^{-6}g/g rock	Th in 10^{-6}g/g rock	Th/U ratio
lividual urements	Mean value			
5 ±0.09	3.9 ±0.5	6.5 ±0.9	17. ±4.	2.6
4 ±0.08				
±0.1	3.2 ±0.4	*3.8 ±0.5	20. ±4.	5.4
±0.3	2.8 ±0.3	3.5 ±0.7	17. ±4.	4.8
±0.1				
±0.2				
0 ±0.06	2.8 ±0.3	6.5 ±1.4	4.3 ±1.0	0.7
0 ±0.07				
±0.1	2.8 ±0.3	2.5 ±0.5	21. ±5.	8.4
±0.2				
±0.2				
±0.1				
0 ±0.05	2.3 ±0.3	.....	.....	...
0 ±0.06				
3 ±0.05				
1 ±0.06				
0 ±0.06				
3 ±0.08	2.1 ±0.3	5.1 ±1.0	2.6 ±0.5	0.5
7 ±0.06				
±0.1	2.0 ±0.4	.....	.....	...
5 ±0.10	1.7 ±0.3	2.7 ±0.5	8.1 ±2.0	3.0
5 ±0.10				
2 ±0.08				
2 ±0.07				
7 ±0.03	1.24 ±0.16	3.5 ±0.7	0.0 ±0.5	0.0
0 ±0.02				
7 ±0.04	0.36 ±0.07	0.34 ±0.06	2.7 ±0.8	7.8
5 ±0.04				
.....	2.3 ±0.2	3.8 ±0.4	10.3 ±1.7	3.7
.....	(11)	(9)	(9)	(9)
±0.1	3.6 ±0.8	4.6 ±0.9	22. ±5.	4.7
±0.1	3.5 ±0.8	3.5 ±0.7	25. ±5.	7.2
±0.1	3.3 ±0.7	3.9 ±0.8	21. ±5.	5.4
5 ±0.03	0.50 ±0.05	.....	.....	...
8 ±0.03				
6 ±0.04				
0 ±0.04				
.....	2.4 ±0.2	3.9 ±0.3	13.4 ±1.6	4.2
.....	(15)	(12)	(12)	(12)

reviously reported radium measurements on 6A and 21A.

an analytical uncertainty of 5 per cent or less, intercomparative analyses between different laboratories may differ by factors of 2 to 100. It is a painful process to discard the results of years of research, and we are naturally reluctant to do so in the light of anything other than the most convincing proof of unreliability. However, in view of the extensive efforts that have been made to avoid all known sources of error in the present work, it seems justifiable to question results by other laboratories when a difference is found to exist, *particularly if it is known that important analytical precautions have not been taken in these laboratories.*

Comparison with former measurements.—A direct comparison of terrestrial radioactivity measurements between laboratories can only be made on identical samples, preferably powdered samples in order to reduce the uncertainty due to inhomogeneity. If a difference is observed, a comparison of standards, in the case of radium and thorium measurements, will aid in isolating the difficulty. An indirect and much less satisfactory comparison between laboratories can be made from a consideration of the average values obtained for the various rock types. This method requires a large number of measurements, and its accuracy is limited by the broad range of radioactivities observed for a given rock type, as is evident from the spread of values given in the tables. For evaluating many of the earlier measurements reported in the literature, only the indirect method is possible, since the samples studied are no longer available. Among investigators still active in the field, a systematic program of interchecking has been in progress for several years, and a number of definite results can be stated at this time.

A co-operative investigation, with two independent methods of measurement, has shown (Evans *et al.*, 1939, p. 931) that all radium determination published by Urry between July 1934 and January 1939 [4] are low by a factor of over two. Between 30 and 40 per cent of this error is explicable by unreliable radium standards, but the remaining source of discrepancy is of an unexplained origin. Because of this uncertainty it does not appear possible to correct Urry's radium measurements by a constant factor. Urry's thorium measurements are also open to slight question, because of analytical uncertainties. Until further work is done to validate these results, it does not appear sound to include them in tabulations of better authenticated radioactivity measurements. For these reasons, extensive research, which otherwise would be an important supplement to our knowledge of terrestrial radioactivity, must be discarded, or at least held in abeyance.

[4] These publications include Lane and Urry (1935, p. 1101-1120), Lane (1935-1938) and Urry (1936a, p. 40-48; 1936b, p. 1218).

TABLE 7.—*Radioactivity measurements of igneous rocks intermediate between acidic and basic*

Sample No.	Rock specimen	Activity index in alphas/hr•mg		U in 10^{-6}g/g rock	Th in 10^{-6}g/g rock	Th/U ratio
		Individual measurements	Mean value			
6I	Hornblende granodiorite	0.39±0.07 0.30±0.07	0.34±0.06	0.82±0.11	0.45±0.08	0.55
4I	La Motte granodiorite	0.80±0.05 0.82±0.05	0.81±0.09	1.6 ±0.3	2.5 ±0.3	1.6
5I	Tholeiite, Cleveland dike	0.7 ±0.1	0.7 ±0.1	1.3 ±0.3	2.7 ±0.4	2.1
7I	Quartz-dolerite, Whin Sill	0.26±0.03	0.26±0.03	0.48±0.11	0.9 ±0.1	1.9
1I	Crescent Hill trachyte	1.8 ±0.1	1.8 ±0.2	2.6 ±0.5	9.7 ±1.1	3.7
2I	Bunsen Peak dacite	1.5 ±0.1	1.5 ±0.15	1.7 ±0.3	10. ±1.	5.9
	Average values for 6 intermediates:		0.90±0.16	1.4 ±0.2	4.4 ±1.2	2.6

TABLE 8.—*Radioactivity measurements of basic igneous rocks*

Sample No.	Rock specimen	Activity index in alphas/hr•mg		U in 10^{-6}g/g rock	Th in 10^{-6}g/g rock	Th/U ratio
		Individual measurements	Mean value			
1B	Porphyritic basalt	1.6 ±0.1	1.6 ±0.2	2.3 ±0.5	8.5 ±1.5	3.7
3B	Crescent Hill basalt	1.1 ±0.1	1.1 ±0.17	1.1 ±0.2	7.8 ±1.5	7.1
7B	Geode Creek basalt	1.1 ±0.1	1.1 ±0.17	0.57 ±0.11	10. ±2.	17.5
10B	Overhanging Cliff	0.70 ±0.10	0.70 ±0.10	0.48 ±0.09	5.9 ±1.0	12.3
37B	Tower Falls basalt	0.58 ±0.05	0.50 ±0.05	.....	.....	....
		0.45 ±0.04				
		0.53 ±0.04				
		0.43 ±0.04				
5B	Columbia River basalt	0.49 ±0.03	0.49 ±0.05	0.88 ±0.06	1.9 ±0.4	2.2
4B	Giant's Causeway	0.44 ±0.03	0.44 ±0.08	1.1 ±0.2	0.45 ±0.08	0.4
6B	Oldwick basalt	0.42 ±0.03	0.42 ±0.05	0.60 ±0.11	2.3 ±0.6	3.8
12B	Basalt	0.40 ±0.07	0.40 ±0.07	0.40 ±0.08	2.9 ±0.5	7.1
	Average values for basalt:	.....	0.75 ±0.09	0.83 ±0.15	5.0 ±0.3	6.8
	Number of specimens:	.....	(9)	(8)	(8)	(8)
14B	Diabase (Horne Mine)	0.51 ±0.05	0.64 ±0.09	1.3 ±0.3	1.8 ±0.4	1.4
		0.77 ±0.06				
$15B_2$	Sudbury olivine diabase	0.61 ±0.04	0.56 ±0.04	0.97 ±0.2	2.4 ±0.3	2.4
$15B_1$	Sudbury olivine diabase	0.55 ±0.05				
		0.53 ±0.05				
$16B_1$	Palisade diabase	0.42 ±0.05	0.39 ±0.02	0.54 ±0.05	2.2 ±0.3	4.0
		*0.44 ±0.04				
$16B_2$	Palisade diabase	0.34 ±0.03				
		*0.41 ±0.04				
$16B_3$	Palisade diabase	0.41 ±0.04				
		*0.37 ±0.03				
$16B_4$	Palisade diabase	0.34 ±0.03				
		0.44 ±0.04				
		*0.33 ±0.03				
$16B_5$	Palisade diabase	*0.41 ±0.04				
18B	Virginia Triassic diabase	0.26 ±0.02	0.32 ±0.03	0.51 ±0.04	1.5 ±0.15	2.9
		0.32 ±0.03				
		*0.38 ±0.04				
	Average values for 4 diabases:	.....	0.48 ±0.05	0.83 ±0.15	2.0 ±0.2	2.7
40B	Gogebic sill	1.86 ±0.07	1.83 ±0.22	1.3 ±0.2	15. ±3.	11.5
		1.80 ±0.11				
38B	Long Lake dike	0.98 ±0.07	0.97 ±0.09	.....	.....	....
		0.78 ±0.05				
		1.10 ±0.06				
		1.00 ±0.07				
$23B_1$	Sudbury trap	0.98 ±0.06	0.83 ±0.08	2.5 ±0.3	0.0 ±0.2	0.0

38B	Long Lake dike	0.98 ±0.07	0.97 ±0.09	.....	.....	....
		0.78 ±0.05				
		1.10 ±0.06				
		1.00 ±0.07				
$23B_1$	Sudbury trap	0.98 ±0.06	0.83 ±0.08	2.5 ±0.3	0.0 ±0.2	0.0
		0.81 ±0.11				
$23B_2$	Sudbury trap	0.69 ±0.04				
$24B_1$	Long Lake trap	0.62 ±0.04	0.67 ±0.07	1.3 ±0.2	2.1 ±0.4	1.6
		0.71 ±0.05				
39B	Gogebic sill	0.70 ±0.05	0.62 ±0.06	0.91 ±0.17	3.2 ±0.3	3.5
		0.59 ±0.03				
		0.75 ±0.05				
		0.57 ±0.04				
25B	Beartooth trap	0.17 ±0.02	0.29 ±0.04	0.60 ±0.08	0.81 ±0.15	1.4
		0.39 ±0.03				
		0.24 ±0.02				
		0.37 ±0.03				
43B	Trap	0.16 ±0.02	0.16 ±0.03	.....	.....	....
41B	Gogebic trap	0.12 ±0.03	0.10 ±0.02	.....	.....	....
		0.10 ±0.04				
		0.08 ±0.04				
42B	Trap	0.043±0.015	0.04 ±0.01	.....	.....	....
	Average values for traps:	.....	0.40 ±0.10	1.3 ±0.2	4.2 ±2.	3.6
	Number of specimens:	.....	(9)	(5)	(5)	(5)
19B	Worthington norite	2.1 ±0.1	2.1 ±0.3	2.2 ±0.3	15. ±3.	6.8
$20B_2$	Sudbury norite	0.94 ±0.09	0.94 ±0.19	1.5 ±0.4	4.3 ±0.8	2.9
21B	Sudbury norite	0.32 ±0.06	0.45 ±0.08	0.71 ±0.14	2.1 ±0.4	3.0
		0.58 ±0.06				
22B	Stillwater norite	0.19 ±0.04	0.22 ±0.03	0.37 ±0.03	0.96 ±0.15	2.6
		0.20 ±0.03				
		0.25 ±0.06				
26B	Riasg Buidhe monchiquite	1.31 ±0.09	1.26 ±0.15	1.9 ±0.4	6.2 ±1.1	3.3
		1.20 ±0.08				
27B	Kilchattan monchiquite	0.54 ±0.05	0.62 ±0.08	0.91 ±0.09	3.3 ±0.6	3.6
		0.66 ±0.06				
32B	Dalhousie andesite	0.45 ±0.08	0.42 ±0.06	1.1 ±0.2	0.45 ±0.7	0.4
		0.38 ±0.06				
28B	Blueberry Mt. gabbro-diorite	0.31 ±0.02	0.31 ±0.04	0.45 ±0.03	1.6 ±0.3	3.6
		0.31 ±0.03				
35B	Watchumna diorite	0.00 ±0.005	0.00 ±0.01	0.00 ±0.03	0.00 ±0.03	....
30B	Dunite, Webster, N. C	0.00 ±0.004	0.00 ±0.01	0.03 ±0.01	0.00 ±0.03	....
	Average values for basic rocks:	.....	0.58 ±0.06	0.95 ±0.09	3.8 ±0.5	4.0
	Number of specimens:	.....	(32)	(27)	(27)	(27)

* Activity index calculated from separate radium and thorium analyses made on these specimens.

An exchange of five powdered rock samples (2A, 14A, 20A, 36A, and 1S) with F. Behounek [5] has clearly established the unreliability of his radium measurements on rocks. Unsystematic errors, ranging up to a factor of 30, were found by this intercheck. Evidence has previously been presented (Evans, 1934, p. 36, 40) which strongly suggests that all of Gräven's (1930, p. 181) and Gräven and Kirsch's (1932, p. 521) terrestrial radioactivity measurements are without foundation. It can also be shown by a simple calculation that the thorium technique described by Hirschi (1920, p. 545) cannot have a sensitivity sufficient to give significant values. Five hand specimens (5A, 2B, 11B, 17B, and 3M), previously analyzed by Joly and Poole, very kindly have been furnished by Professor Poole for intercheck measurements. The radium measurements on these rocks, reported in Tables 1, 3, and 4, are invariably lower than the earlier values of Joly and Poole, given in the geologic notes. It will be seen by a comparison of the two sets of values for these five specimens that no fixed ratio obtains. However, Joly and Poole's results show a greater ratio to the present values for the specimens with lower radium content. This observation suggests the possibility of a systematic error in the earlier work which may be due to radioactive contamination in the chemicals used by Joly and Poole. The recent independent agreement on the radioactivity of sea water between Føyn and his coworkers (1939, p. 10) and Evans *et al.* (1938, p. 243) also furnishes additional evidence that Joly's radium values, on all types of specimens, are systematically too high. It appears that the measurements of a number of other workers (Evans *et al.*, 1938, p. 243, Table I) are also subject to question as the result of these researches. The present international situation has interfered with further interchecking with European laboratories, but it is hoped that in the not too distant future these differences can be studied in greater detail.

An exchange of a number of rock specimens has also been made with the Geophysical Laboratory in Washington, D. C. Radium measurements on four rock specimens (2A, 14A, 20A, and 36B) show ratios between the earlier values of Piggot, given in the geologic notes, and the present values, reported in Tables 1 and 3, of 1.4 to 2.8, with an average of very close to 2. Intercheck measurements on the standard solutions used for calibration are in good agreement, indicating that these solutions are not the source of the differences. More recent radium measurements by Urry, working in Piggot's laboratory, using the 12 standard rock samples (*see* 3A in geologic notes) for comparative measurements, are in quite good agreement with the values reported for these samples in Tables

[5] Formerly Director of the Radiological Institute of Czechoslovakia.

1, 3, and 5. Further work is in progress to resolve the disagreement on the earlier specimens.

On the other hand sufficient intercheck measurements have been made on the same rock samples among Evans, Clarke, Finney, Goodman, Keevil, and Raitt to establish the mutual agreement of the results summarized in Table 9 [6] and those presented in the preceding tables. These data are combined in Table 10 to give more representative average values for the different rock types.

A number of summaries of the radioactivity of terrestrial materials have previously appeared,[7] but the emphasis has invariably been on the geological implications with little or no consideration of the probable reliability of the measurements themselves. As a result many of the geological conclusions suffer from the lack of critical evaluation of the data on which they are based.

Jeffreys' (1936, p. 149-170) compilation of the radioactivities of igneous rocks is the most complete summary available. The measurements of Evans and his coworkers, given in Table 9, and omitted from Jeffreys' summary, would lower Jeffreys' averages to a slight extent. Other data not included are those of Urry, which were omitted because they appeared anomalously low compared to other observations. As indicated above, we now know that these results are erroneously low. Intercheck measurements with other workers whose results are included in Jeffreys' averages suggest that where other errors exist they are very likely in the direction to give unreliably high values. For these reasons a comparison of the weighted average values of Table 10 with Jeffreys' average values gives a rather reliable indication of the downward revision that appears necessary in estimates of the radioactive content of igneous rocks (Table 11). Too few specimens of intermediate rocks have been measured to give conclusive results. However, since a definite decrease in the average radioactivities is indicated for both the acidic and basic rocks, a similar suggested change in the intermediates lends support to the significance of the observed differences. A decrease of 40 per cent in the estimated average radium content of acidics and a 60 per cent decrease for basics are indicated in Table 11. Decreases of 25 and 50 per cent in the estimated average thorium contents are also indicated for acidics and basics respectively. The decreases in both the radium and thorium values are

[6] In addition to the results included in Table 9, Keevil (1938a, p. 146; 1938b, p. 690-691) has published a number of other measurements. These analyses were made on rock specimens also measured by the authors and included in Tables 1 to 8. Because inclusion of Keevil's duplicate analyses in Table 9 would give undue weight to certain specimens in the averages of Table 10, his results have been omitted.

[7] Among others are: Holmes (1914, p. 12; 1915, p. 2; 1926, p. 1055; 1931, p. 559; 1937, p. 763), Hoffmann (1939, p. 189), Hatuda (1936, p. 1), Jeffreys (1936, p. 149), Poole and Joly (1924, p. 819). Piggot (1931, p. 28), and Urry (1933, p. 125).

TABLE 9.—*Contemporaneous radioactivity measurements of rocks*

Sample No.	Rock specimen	Ra content in 10^{-12} g/g rock	Activity index in alphas/hr·mg	Uranium in 10^{-6} g/g rock	Thorium in 10^{-6} g/g rock	Th/U ratio
C-6	Granite, Churchill, Manitoba	0.23	1.00	0.62	8.7	14.
C-7	Red granite, Pike's Peak	1.52	5.77	4.1	48.0	12.
C-19	Lava, Arequipa, Peru	1.75	5.70	4.7	44.5	9.5
C-20	Lava, Crucero Alto, Peru	2.38	2.55	6.4	1.4	0.2
A-3	Quartz monzonite, Kaweah Quad., Calif	0.47	1.46	1.3	11.	8.5
A-5	Granite, Kaweah Quad., Calif	0.53	1.97	1.5	16.	11.
A-6	Granite, Rocky Hill Quad., Calif	0.59	1.93	1.7	15.	8.8
A-7	Quartz monzonite, Raymond, Calif	0.41	0.77	1.2	3.9	3.3
Gal (?)	Granite, Gananoque, Ont	0.37	0.63	1.05	2.82	2.7
C1	Granite, E. La Motte Township, Ont	0.44	0.64	1.25	2.04	1.6
T2A(?)	Granite, East Taschereau, Que	0.56	0.97	1.59	4.4	2.8
T2A(?)	Granite, West Taschereau, Que	0.20	0.29	0.58	0.9	1.5
KF3	Granite, Kingston, Ont	0.49	0.79	1.4	3.1	2.3
NB5	Granite, Hampstead, N. B	1.55	3.12	4.40	17.0	3.9
	Average values for 14 acidic rocks:	0.82±0.12	2.0 ±0.3	2.3 ±0.3	12.8 ±2.7	5.9
1	Basalt, Mt. Harkness	0.30	....	....	....	....
2	Basalt, NE of Butte Lake	0.35	....	....	....	....
5	Andesite, Raker Peak	0.36	....	....	....	....
6	Andesite, Brokeoff Volcano	0.28	....	....	....	....
7	Andesite, Badger Mt	0.60	....	....	....	....
8	Dacite pumice, Lassen Peak	0.80	....	....	....	....
9	Andesite, Crystal Lake	0.70	....	....	....	....
10	Dacite, Bumpass Hell	0.58	....	....	....	....
11	Dacite (1915), Lassen Peak	0.70	....	....	....	....
12	Dacite, Lassen Peak	0.81	....	....	....	....
13	Dacite, Chaos Crag	0.80	....	....	....	....
14	Dacite, Raker Peak	1.14	....	....	....	....
A-1	Quartz diorite, Kaweah, Quad., Calif	0.87	1.79	2.5	10.	4.0
A-2	Quartz diorite, Lemon Grove Quad., Calif	0.37	1.07	1.1	7.7	7.0
(*)	Val Verde tonalite	0.46	0.93	1.3	5.1	3.8
113	Basalt, Eustis Mine, Que	0.22	0.37	0.62	1.61	2.6
NB2	Basalt, St. George, N. B	0.17	0.33	0.49	1.70	3.5
37	Basalt, Gogebic Range, Wis	0.14	0.30	0.40	1.72	4.3
R	Basalt, Ropruchei, U. S. S. R	0.27	0.46	0.77	2.04	2.7
	Average value for basic rocks:	0.52±0.03	0.75±0.13	1.0 ±0.2	4.3 ±0.9	4.0
	Number of specimens:	(19)	(7)	(7)	(7)	(7)
C-11	Quarry limestone, Minneapolis, Minn	0.60	0.87	1.6	3.0	1.9
C-16	Limestone, Cormorant Lake, Manitoba	0.11	0.13	0.30	0.20	0.7
C-17	Limestone, Riverside, Calif	0.32	0.34	0.86	0.16	0.2

113	Basalt, Eustis Mine, Que	0.22	0.31	0.62	1.61	2.6
NB2	Basalt, St. George, N. B	0.17	0.33	0.49	1.70	3.5
37	Basalt, Gogebic Range, Wis	0.14	0.30	0.40	1.72	4.3
R	Basalt, Ropruchei, U. S. S. R	0.27	0.46	0.77	2.04	2.7
	Average value for basic rocks:	0.52±0.03	0.75±0.13	1.0 ±0.2	4.3 ±0.9	4.0
	Number of specimens:	(19)	(7)	(7)	(7)	(7)
C-11	Quarry limestone, Minneapolis, Minn	0.60	0.87	1.6	3.0	1.9
C-16	Limestone, Cormorant Lake, Manitoba	0.11	0.13	0.30	0.20	0.7
C-17	Limestone, Riverside, Calif	0.32	0.34	0.86	0.16	0.2
C-18	Kaibab limestone, Grand Canyon, Colo	0.95	1.15	2.6	2.0	0.8
C-22	Limestone, Cormorant Lake, Manitoba	0.12	....	0.33	....	....
	Average values for limestones:	0.42±0.11	0.6 ±0.2	1.3 ±0.7	1.1 ±0.3	0.9
	Number of specimens	(5)	(4)	(5)	(5)	(4)
C-1	Roadway gravel	0.41	....	1.1	....	....
C-2	Roadway gravel	0.25	....	0.68	....	....
C-3	Glacial rubble	0.33	....	0.90	....	....
C-5	Gravel	0.52	1.26	1.4	8.3	5.9
C-10	Pit gravel, Minneapolis, Minn	0.48	0.82	1.3	3.8	2.9
C-4	Glacial sand	0.44	....	1.2	....	....
C-12	Sandstone, Zion Canyon, Utah	0.32	0.80	0.86	5.4	6.3
C-21	Quartzite, Crucero Alto, Peru	0.20	....	0.54	....	....
C-14	Soil, Ellendale, N. Dakota	0.78	....	2.1	....	....
C-15	Soil, Spokane, Wash	0.68	....	1.8	....	....
	Average values for sedimentary rocks:	0.44±0.04	0.8 ±0.1	1.2 ±0.1	3.3 ±0.7	2.7
	Number of specimens:	(15)	(7)	(15)	(7)	(7)

* Samples 730, 750, 811, 830, 930, and 953 of tonalite batholith from Val Verde tunnel, located 13 miles southeast of Riverside, California. Average values of radioactivity as determined by Clarke (1937, p. 64); small corrections have been applied for more recent values of disintegration constants.

Note: C-series samples Ra by Evans and Raitt (1935, p. 172); activity index by Raitt (1935, p. 71), Th calculated according to Eq. 1.

A-series samples measurements by Finney (1935, p. 70).

Samples No. 1-14 by Evans and Williams (1935, p. 446).

Other samples by Keevil (1938a, p. 146; 1938b, p. 690–691), second classification of sample NB2 accepted.

TABLE 10.—*Summary of average radioactivities*

Rock type	Tables	Ra content in 10^{-12} g/g rock	Activity index in alphas/hr•mg	Uranium in 10^{-6} g/g rock	Thorium in 10^{-6} g/g rock	Th/U ratio
Acidic igneous	1, 6	1.64±0.20 (29)	2.4 ±0.2 (15)	3.9 ±0.3 (12)	13.4 ±1.6 (12)	4.2 (12)
Acidic igneous	9	0.82±0.12 (14)	2.0 ±0.3 (14)	2.3 ±0.3 (14)	12.8 ±2.7 (14)	5.9 (14)
Acidic igneous (weighted average):		1.37±0.17 (43)	2.2 ±0.2 (29)	3.0 ±0.3 (26)	13. ±2. (26)	5.0 (26)
Intermediate igneous	2, 7	0.51±0.05 (7)	0.90±0.16 (6)	1.4 ±0.2 (6)	4.4 ±1.2 (6)	2.6 (6)
Basic igneous	3, 8	0.31±0.03 (35)	0.58±0.06 (32)	0.95±0.09 (27)	3.8 ±0.5 (27)	4.0 (27)
Basic igneous	9	0.52±0.03 (19)	0.75±0.13 (7)	1.0 ±0.2 (7)	4.3 ±0.9 (7)	4.0 (7)
Basic igneous (weighted average):		0.38±0.03 (54)	0.61±0.07 (39)	0.96±0.11 (34)	3.9 ±0.6 (34)	4.0 (34)
Sedimentary	5	0.71±0.12 (13)	.....	.....	.....	....
Sedimentary	9	0.44±0.04 (15)	0.8 ±0.1 (7)	1.2 ±0.1 (15)	3.3 ±0.7 (7)	2.7 (7)
Sedimentary (weighted average):		0.57±0.08 (28)	.....	.....	.....	....

( ) Number of rock specimens on which average value is based.

TABLE 11.—*Comparison of average radioactivities*

Rock type	Average radium in 10^{-12} g/g rock		Average thorium in 10^{-6} g/g rock		Average Th/U ratio	
	Present Estimate	Jeffreys' Estimate	Present Estimate	Jeffreys' Estimate	Present Estimate	Jeffreys' Estimate
Acidic igneous	1.37±0.17 (43)	2.38±0.16 (122)	13. ±2. (26)	17.6 ±2.2 (60)	5.0 (26)	2.6 (60)
Intermediate igneous	0.51±0.05 (7)	1.85±0.25 (36)	4.4 ±1.2 (6)	18.9 ±8.5 (4)	2.6 (6)	3.6 (4)
Basic igneous	0.38±0.03 (54)	1.03±0.05 (122)	3.9 ±0.6 (34)	7.8 ±0.6 (77)	4.0 (34)	2.7 (77)

Present estimates from Table 10; Jeffreys' estimate from his compilation (Jeffreys, 1936, p. 150).
( ) Number of rock specimens on which average value is based.

hardly surprising since Jeffreys' averages are strongly dependent upon the measurements of Joly and Poole, Gräven and Kirsch, and Hirschi, all of which appear to be on the high side.

Possible sources of error.—One interpretation of the larger percentage decrease indicated for basic rocks as compared with acidic rocks is that the mean errors in the earlier radium and thorium determinations were fairly constant. Under these conditions, the error would contribute proportionately less to the more radioactive (and, therefore, more readily measurable) acidic rocks than to the less radioactive basic rocks. Contamination errors are of this nature. If a reliable correction factor could be found, it might be possible to salvage a significant fraction of the earlier work in this field.

The indicated increase in Th/U ratio may be the result of proportionately larger contamination errors in the earlier radium measurements than in those of thorium. As yet, there is no definite proof of this point, but Føyn *et al.* (1939, p. 40) have observed a greater likelihood of radium contamination than uranium and thorium in ordinary chemical reagents.

General discussion.—It was observed quite early (Strutt, 1905, p. 472) that the concentration of radioactive elements in rocks increases with silica content. A number of other investigations in addition to the present work have amply confirmed this general conclusion. A correlation of radium with potassium content has been reported (Evans and Williams, 1935, p. 447; Hatuda, 1936, p. 15). A notable exception to this latter observation is the lack of correlation with potassium but the apparent correlation with biotite content observed by Barth (1938, p. 240) in the Finnish granites measured by Piggot (1938, p. 227). The principal difficulty with most of these investigations is that the radioactivity measurements have been made on the composite rock instead of on the separate mineral constituents. In addition, two aspects of this problem seem to have escaped most workers in this field. The first, and rather obvious consideration, is that, while radium is the substance usually measured, the parent element, uranium, must be the one concentrated in the rock at the time of its solidification in order for radium and the other elements in the series to be present today, except for very recently formed rock bodies. The second point is that there is no particular reason to expect the same correlation to apply to rocks of totally different genesis, even though they may be of the same general chemical composition. Certainly the conditions of crystallization were entirely different for the Lassen lavas (Evans and Williams, 1935, p. 441) as compared with those that obtained when the Finnish granites (Barth, 1938, p. 231) were formed.

Since it now appears necessary to base helium age measurements on selected mineral constituents of rocks rather than on the rocks as a whole (Hurley and Goodman, 1941, p. 553), an increasing number of radioactivity measurements on separate minerals will probably appear as an important by-product of age researches in the future. These results should be of considerable value in solving the geochemical problems of the distribution of radioactive elements in rocks.

RADIOACTIVE HEAT

GENERAL STATEMENT

The radiations emitted by the radioactive elements contained in the earth are absorbed by the rocks, and the energy of disintegration appears in the form of heat. Since radioactivity is a nuclear property of atoms, the rate of decay would not be expected to be affected by changes of pressure, temperature, or chemical composition—even such high temperatures and pressures as must exist deep in the earth. There is considerable experimental evidence to support this conclusion.

The energies of the three types of radiation emitted by the members of the uranium, actino-uranium, and thorium series have been accurately measured. From these data and the disintegration constants of the parent radioactive elements, the specific rate of generation of heat for each radioactive series (in equilibrium) can be computed as shown below.

DERIVATION OF CONVERSION FACTORS

The energy accompanying the emission of alpha radiation constitutes the source of more than 85 per cent of the heat generated by the naturally occurring radioactive elements. The alpha-ray energies for the UI and Th series, including the correction for the recoil nucleus, are summarized in Table 12. Combining the total values for these series with the following accepted physical constants.[8]

$\lambda_{UI} = 4.87 \times 10^{-18}$ sec^{-1}, $\lambda_{Th} = 1.58 \times 10^{-18}$ sec^{-1}
Atomic weights: U = 238.14, Th = 232.12
Avogadro's number: $N = 6.02 \times 10^{23}$ atoms per gram equivalent
1 calorie $= 4.19 \times 10^{7}$ ergs, 1 MEV $= 1.60 \times 10^{-6}$ ergs, 1 year $= 3.15 \times 10^{7}$ sec.

the heating effect for the radiations can be calculated as follows:

Ratio (Rutherford *et al.*, 1930, p. 161) $\alpha : \beta : \gamma = 124.8 : 6.3 : 9.4$

$$\text{hence: } \frac{\alpha + \beta + \gamma}{\alpha} = \frac{140.5}{124.8} = 1.125$$

[8] λ_{UI} from Kovarik and Adams (1941) including Nier's (1939, p. 150) value for the actino-uranium /UI activity ratio. λ_{Th} from Kovarik and Adams (1938, p. 413).

Therefore, heat per gram UI per year =

$$\frac{43.34 \times 1.125 \times 1.60 \times 10^{-6} \times 3.15 \times 10^{7}\ N\ \lambda_{UI}}{4.19 \times 10^{7}\ UI} = 0.708 \text{ calories}$$

Heat per gram U per year = 0.741 calories[9]

Similarly, heat per gram Th per year =

$$\frac{36.82 \times 1.125 \times 1.60 \times 10^{-6} \times 3.15 \times 10^{7}\ N\ \lambda_{Th}}{4.19 \times 10^{7}\ Th} = 0.203 \text{ calories}$$

TABLE 12.—*Uranium and thorium series alpha-ray energies*

Element	Mean range in cm In air at 15°C 760 mm Hg	Disintegration energy in million electron volts
UI	2.65	4.13
UII	3.21	4.75
Io	3.09	4.66
Ra	3.26	4.87
Rn	4.05	5.58
Ra A	4.66	6.11
Ra C′	6.91	7.83
Ra F	3.84	5.41
Total UI series		43.34
Th	2.60	4.31
RdTh	3.95	5.47
ThX	4.28	5.78
Tn	5.00	6.39
Th A	5.64	6.90
35% Th C (av.)	4.73	2.16
65% Th C′	8.57	5.81
Total Th series		36.82

The heating effect of potassium is estimated from Mülhoff's (1930, p. 205) determination of the specific activity as 23(± 15 per cent) beta rays per second per gram of K and values of 0.7 MEV for the maximum beta ray continuous spectrum (Anderson and Neddermeyer, 1934, p. 653) and 2 MEV for the gamma ray energies (Mülhoff, 1930, p. 205; Gray and Tarrant, 1934, p. 694). The latter investigators have shown that three gamma ray quanta are emitted per 100 disintegration electrons. Combining these data, the heat per gram of K per year $= 4.6 \times 10^{-6}$ calories. Taking into account the uncertainties in the physical quantities, the rate of heat generation for the three most abundant naturally occurring radioactive substances can be summarized as:

(UI + AcU) Series: 0.74 ± 0.03 cal/yr/gm U

Th Series: 0.20 ± 0.01 cal/yr/gm Th

Potassium: $5 \pm 2 \times 10^{-6}$ cal/yr/gm K

[9] Addition of 4.6 per cent for AcU series (Nier, 1939, p. 150).

HEAT GENERATION IN ROCKS

Using the above conversion factors and the average radio-active content of rocks from Table 10, an estimate of the rate of generation of heat in the different types of rocks can be made. The resulting values, including the relatively small contribution from potassium,[10] are compared with a similar earlier estimate by Holmes (1930, p. 136; 1931, p. 559) in Table 13.

TABLE 13.—*Estimates of radioactive heat generation in rocks*

	Heat production in 10^{-6} calorie per gram rock per year			
	Acidic igneous	Intermediate igneous	Basic igneous	Sedimentary
(Holmes' values*)				
U.....	7.1	6.1	2.5	....
Th.....	4.6	4.1	1.5	....
K.....	4.2	3.1	1.7	....
Total.....	15.9	13.3	5.7	....
(Present values)				
U.....	2.2 ±0.2	1.0 ±0.15	0.7 ±0.08	0.9 ±0.09
Th.....	2.6 ±0.4	0.9 ±0.2	0.8 ±0.01	0.7 ±0.1
K.....	0.14±0.02	0.10±0.02	0.07±0.01	0.10±0.02
Total.....	5.0 ±0.6	2.0 ±0.4	1.6 ±0.1	1.7 ±0.2

* Holmes' classification of igneous rocks into the three broad types differs somewhat from that used in the present discussion. Holmes' values have been rearranged according to the present grouping of rocks as indicated in Tables 1 to 8.

The extent to which Holmes' values have been generally adopted is reflected by their inclusion, in part or as a whole, in numerous publications (Daly, 1933, p. 70; Grout, 1932, p. 146; Ellsworth, 1932, p. 65; Hevesy and Paneth, 1938, p. 274). The estimates based on the present values for the radioactive content and the conversion factors are considerably lower than those of Holmes. These differences are largely attributable to (1) the lower estimates for the radioactivities and (2) a decrease in the estimated geothermal importance of potassium as the result of more precise studies of the radioactive properties of this element in recent years.

CONCLUSIONS

Several hundred radioactivity measurements have been made on rocks using unusual analytical care. The results of these measurements have been combined with the limited number of well-authenticated results

[10] The potassium content has been estimated from average chemical analyses reported by Daly (1933, p. 9-28) and Grout (1932, p. 127) to be: $2.8 \pm 0.5 \times 10^{-2}$g/g for acidic igneous rocks, $2.0 \pm 0.4 \times 10^{-2}$g/g for intermediate igneous rocks, $1.4 \pm 0.3 \times 10^{-2}$g/g for basic igneous rocks, and $2.0 \pm 0.4 \times 10^{-2}$g/g for sedimentary rocks.

previously reported by other investigators to obtain estimated average values for broad lithologic types. When compared with previous averages, the present values indicate that a substantial downward revision is necessary in the present conception of the radioactive content of igneous rocks. Geological considerations based on the earlier values will very likely need revision. In particular Holmes' estimates of the rate of generation of radioactive heat within the earth may need to be substantially decreased.

GEOLOGIC NOTES ON ROCK SPECIMENS REFERRED TO IN TABLES

Acidic Igneous Rocks

1A Brifosse granite obtained together with 22A from E. Rothe for intercheck radioactivity measurements. Gamma-ray activity of these rocks reported by Rothe and Hopcewicz (1937, p. 165) and by Rothe and Hee (1937, p. 1835).

2A Kumlinge granite, Aland Archipelago, submitted as a powder by C. S. Piggot, one of four specimens (others: 14A, 20A, and 36B) for intercheck measurements. C. S. P.'s mean value. 5.51×10^{-12} g Ra/g, and geologic notes previously reported (Piggot, 1938, p. 228) using designation P-75.

3A Missourian granite 50-pound specimen collected by U. S. Geological Survey from the Sheahan Quarry, in about Section 14, T. 34 N., R. 3 E., Graniteville, Missouri. One of 12 rocks (others: $4A_2$, 19A, 5B, 8B, 18B, 28B, 30B, 31B, 5S, 9S, and 11S) being used as standard samples for the international intercalibration of radioactivity measurements (Goodman and Evans, 1940, p. 557). This specimen was erroneously designated (Curtiss *et al.*, 1940, p. 602) as "Carthage" granite.

$4A_1$ Chelmsford granite collected by N. B. Keevil from H. E. Fletcher Co. quarry, N. Chelmsford, Mass. Age: possibly Mississippian (Emerson, 1917).

$4A_2$ Chelmsford granite, 50-pound specimen collected by C. Goodman from same location as $4A_1$. One of standard samples; *see* 3A.

5A Rapakiwi granite from Viborg, Finland, one of five hand specimens (others: 2B, 11B, 17B, and 3M) submitted for intercheck radium measurements by J. H. J. Poole. These specimens are particularly valuable since they not only furnish an intercheck with the recent work of Poole (1938, p. 595) but also afford a means of evaluating the earlier results of the late J. Joly, since some of the rocks were measured by the latter. Poole (personal communication, 1938) reports Joly's value as 5.77×10^{-12} g Ra/g for 5A.

6A Granite collected by R. A. Millikan from boulder in Glen Cove, Colorado. *See* Evans and Raitt (1935, p. 172) and Raitt (1935) for other radioactivity measurements, designation C-13.

7A Rockport granite, Lat. 42° 40′ N, Long. 70° 35′ W., collected by N. B. Keevil from Upper Pigeon Hill Quarry, Cape Ann, Mass. Intrudes Salem gabbro diorite, which in turn cuts pre-Cambrian rocks and is possibly of Devonian age. It is cut by diabase dikes interbedded with Triassic sedimentary beds at Deerfield and Mt. Holyoke, Mass. Age: Probably pre-Triassic and post-Cambrian.

8A Rockport xenolith, fine-grained and cognate, from the same large hand specimen as 7A.

9A Murray granite collected by H. C. Cooke, 1939, from railway cut, 4 to 5 feet from surface, lot 10, range V, McKim Township, Sudbury, Ont.

$10A_1$ Creighton granite collected by H. C. Cooke, 1939, from road cut 5 feet from surface, SW quarter, lot 9, concession 1, Snider Township, Sudbury, Ont.

$10A_2$ Creighton granite collected by E. A. Collins from surface rock SE of Mud Lake, Copper Cliff, Ont. Considered to be outcrop of $10A_3$.

$10A_3$ Creighton granite collected by E. A. Collins from the Creighton Mine, 40 level, 14 X-cut, Copper Cliff, Ont.

11A Granite collected by N. B. Keevil from quarry in Franklin, Me.; *see* Dale (1923, p. 219). Age: Middle Paleozoic.

$12A_1$ Quincy granite collected by N. B. Keevil from J. S. Swingle quarry, Quincy, Mass. Age: Possibly Mississippian, probably late Paleozoic.

$12A_2$ Quincy granite drill core from over 600 feet deep, same location as $12A_1$, obtained from R. A. Daly.

13A Granite from collection of G. D. Louderback. Late Jurassic from the Sierra Nevada, Mokelumne River region, quarry for dam.

14A Granite collected by J. J. Sederholm and T. F. W. Barth in summer of 1932 from Ava, north of Brando, Aland Archipelago. Submitted as a small hand sample by C. S. Piggot for intercheck measurements; *see* 2A. Piggot's mean value, 1.21×10^{-12} g Ra/g, and geologic notes previously reported (Piggot, 1938, p. 229) using designation P-83.

15A Fitchburg granite collected by N. B. Keevil from Rollstone Hill quarry, Fitchburg, Mass. Age: probably Carboniferous.

16A Granite from Kawatana, H. S., Japan submitted by Z. Hatuda together with three other specimens (18A, 27A, and 1M) for intercheck radioactivity measurements.

17A Granite collected by N. B. Keevil from Barryfield quarries, 2 miles east of Kingston, Ont. Age: post-Grenville and pre-Keweenawan.

18A Granite from Azi, collection of Z. Hatuda; *see* 16A.

19A Granite, 50-pound specimen obtained by C. Goodman indirectly from H. E. Fletcher Co. quarry in Milford, Mass. One of standard rock samples; *see* 3A. Compact, massive rock, somewhat above medium grain and of light color. Its two especially characteristic constituents constantly present are blue quartz and a microcline-microperthite. Age: post-Cambrian and pre-Carboniferous.

20A Granite from Ounasjok in Rovaniemi, end of the railway in northern Finland, collected by J. J. Sederholm and T. F. W. Barth in the summer of 1932. Submitted as a small hand specimen by C. S. Piggot for intercheck measurements; *see* 2A. Piggot's mean value, 0.43×10^{-12} g Ra/g, and geologic notes previously reported (Piggot, 1938, p. 229) using designation P-86.

21A Granite collected by R. A. Millikan from outcrop in Red River, Winnipeg, Manitoba. *See* Evans and Raitt (1935, p. 172) and Raitt (1935) for other radioactivity measurements, designation C-8.

22A Rhyolite du Rosskopf obtained from E. Rothe for intercheck radioactivity measurements; *see* 1A.

23A Rhyolite collected by A. D. Howard 100 feet north of Tower Creek Bridge, Yellowstone Park.

24A Rhyolite collected by C. Max Bauer from Golden Gate, Yellowstone Park.

25A Porphyritic rhyolite collected by C. Max Bauer from near 8-mile Bridge, Yellowstone Park.

26A Late aplite phase of Creighton granite, 10A, collected by H. C. Cooke, 1939, from 4 feet below original surface, lot 1, concession III, Snider Township, Sudbury, Ont.

27A Pitchstone from Aso volcano, collection of Z. Hatuda, *see* 16A.

28A Obsidian collected by C. Max Bauer from Obsidian Cliff, Yellowstone Park.

29A Micropegmatite collected by H. C. Cooke, 1939, from railway cut 6 feet below surface, lot 1, concession III, Wisner Township, Sudbury, Ont.

30A Granite from Mt. Manitou, Colo., collection of R. A. Millikan. *See* Evans and Raitt (1935, p. 172) and Raitt (1935) for other radioactivity measurements, designation C-9.

31A Lebanon granite collected by E. P. Kaiser (1938, p. 107) near Hanover, N. H.

32A Granite collected by N. B. Keevil of unknown location, designation K-G-1.

33A Pegmatite sample collected by A. C. Lane, 1937, from Ruggles Mine, Grafton, N. H.

BASIC IGNEOUS ROCKS

1B Porphyritic basalt collected by A. D. Howard from SE base of Cresent Hill, Yellowstone Park.

2B Vesicular basalt from Gravenoire volcano, Clermont, France, submitted as a small hand specimen for intercheck radioactivity measurements by J. H. J. Poole who reports 1.19×10^{-12} g Ra/g as Joly's value (personal communication, 1938); *see* 5A.

3B Cresent Hill basalt collected by A. D. Howard ¼ mile west of crossing of Geode Creek, Yellowstone Park.

4B Basalt from the well-known Giant's Causeway in Ireland, submitted as a large hand specimen for intercheck measurements by Arthur Holmes (1937, p. 155); *also see* 5I and 7I.

5B Columbia River basalt, 50-pound specimen secured through A. W. Fahrenwald. One of standard samples; *see* 3A. Composed of calcic labradorite, augite, olivine, and a little magnetite. Age: Middle Miocene.

6B Basalt from Lat. 40° 40′ N., Long. 40° 45′ W., collected from a flow by A. C. Hawkins, 1936, 50 feet from the surface in a working quarry at Oldwick, N. J. The top of the flow has been removed by erosion, and the bottom is not visible. Similar flows in the area, 16B and 17B, of slightly different periods of eruptivity have been identified as being of Triassic age from fossils in adjoining sedimentary beds (red shales and sandstones). Age: Probably Triassic.

7B Basalt collected by A. D. Howard ¾ mile E of crossing of Geode Creek, Yellowstone Park.

8B Deccan Trap Plateau basalt, 50-pound specimen obtained from near Bombay, India, through the courtesy of the U. S. Geological Survey and A. S. Kalapesi, St. Xaviers College. One of standard samples; *see* 3A. Composed of plagioclase (calcic andesine), augite (roughly 50 per cent), less magnetite, and a small amount of secondary minerals including chloro-phalite-delessite.

$9B_1$ Second Watchung trap collected Dec. 7, 1939, by P. M. Hurley from old quarry on Murray Hill short distance SW of Summit, N. J., on top of Second Watchung Mtn. Specimen from 10 feet above base of a flow at least 30 feet thick and taken about 25 feet below original surface. Rock has fine diabasic texture.

$9B_2$ First Watchung trap collected Dec. 7, 1939, by P. M. Hurley from fresh blast in operating quarry on First Watchung Mtn. opposite Summit, N. J., about 1 mile east of Summit. Specimen from block of fresh broken rock probably coming from a point in the face between 30 and 50 feet below the original surface. No sign of limonite on any of the material.

10B Basalt collected by A. D. Howard from Overhanging Cliff, Yellowstone Park.

11B Basaltic lava from Hawaiian Islands submitted as a small hand specimen for intercheck radioactivity measurements by J. H. J. Poole who reports 1.25×10^{-12} g Ra/g as Joly's value (personal communication, 1938); *see* 5A.

12B Basalt collected by C. Max Bauer from near 8-mile Bridge, Gardiner, Yellowstone Park.

13B Oxbow Creek basalt collected by A. D. Howard ¼ mile west of crossing of Oxbow Creek, Yellowstone Park.

14B Diabase from Lat. 43° 15′ N., Long. 79° 2′ W. collected by P. Price 3 feet from chilled contact of a N-S dike at the 17th level (1975 ft.) of the Horne Mine, Norando, Quebec. In a previous publication (Evans *et al.*, 1939, p. 946) this rock was indicated as post-Cobalt in age. Subsequently, a personal communication from H. C. Cooke informs us that this dating is very uncertain and the most that can be said is that this rock is post-Timiskaming. (*See* Geological Survey of Canada Memoir 166, p. 143-144.)

$15B_1$ Olivine diabase (coarse-grained phase) collected by E. A. Collins from hanging wall of pit at fault, Creighton Mine, Copper Cliff, Ont. Considered to be outcrop of $15B_2$.

$15B_2$ Olivine diabase collected by E. A. Collins from the Creighton Mine, 20 level, Main No. Dr., Copper Cliff, Ont.

$15B_3$ Olivine diabase collected by H. C. Cooke, 1939, from about 2 feet below original surface, west side of lot 10, concession V, McKim Township, Sudbury, Ont.

$16B_1$ Palisade diabase collected Dec. 8, 1939, by P. M. Hurley from old quarry on Forest Ave., Staten Island. Specimen from 8 feet below original surface—fresh hard diabase, surface glaciated, and no weathering.

$16B_2$ Palisade diabase collected by P. M. Hurley, Dec. 8, 1939, from road cut on River Road at corner of 16th St., North Bergen, N. J. Specimen is a fine-grained trap, 20 feet below original surface. This point is the south end of the main Palisades.

$16B_3$ Palisade diabase collected by P. M. Hurley, Dec. 8, 1939, from large old quarry by side of River Road facing the Hudson, 1 mile north of $16B_2$, near Guttenberg, N. J. Specimen of dense trap, 150 feet below original surface, possibly close to bottom of sill.

$16B_4$ Palisade diabase collected, Dec. 8, 1939, by P. M. Hurley from road cut on State highway 5 (N.J.) as it winds up the Palisade bluff above the 125th St. Ferry, Edgewater. Specimen from 70 feet below top of bluff and probably at least 30 feet from original surface.

$16B_5$ Palisade diabase collected Dec. 6, 1939, by P. M. Hurley from Kingston Trap Quarry 1½ miles from Kingston, N. J., on road toward Rocky Hill, from a fresh blast, about 20 feet original surface; quarry operating.

17B Diabase from Somerset Co., N. J., submitted for intercheck measurements by J. H. J. Poole, who reports (personal correspondence, 1938) Joly's value as 1.18×10^{-12} g Ra/g; *see* 5A.

18B Triassic diabase collected through the courtesy of the U. S. Geological Survey from Centerville, Va. Composed of calcic labradorite, about 40 per cent augite, a very little magnetite, and sparse chloritic and serpentine alteration products. Rock very fresh; grain size about 0.5 to 1.0 mm.

19B Norite collected by E. M. Kindle from nickel-norite offset dike rock, Worthington Mine, railway cut about 100 feet from Worthington Station, Ont.

$20B_1$ Norite collected by H. C. Cooke, 1939, from railway cut 15 feet below surface, lot 1, concession III, Wisner Township, Ont.

$20B_2$ Norite collected by E. A. Collins from hanging wall of pit at fault, Creighton Mine, Copper Cliff, Ont. Considered to be outcrop of 21B.

21B Norite collected by E. A. Collins from Creighton Mine, 40 level, 14 X-cut, Copper Cliff, Ont.

22B Norite, Lat. 45° 1′ N., Long. 109° 25′ W., collected July 1937 by A. C. Lane from the metamorphosed Stillwater complex of Quad Creek, Mont., 1 mile north of State line. The Stillwater complex is bordered by a Cambrian conglomerate at an erosional contact and is cut by granite and by basic dikes. Age: Probably early pre-Cambrian.

$23B_1$ Trap collected by E. A. Collins from the Creighton Mine, 40 level, 14 X-cut, Copper Cliff, Ont.

23B$_2$ Trap collected by E. A. Collins from the hanging wall of pit at fault, Creighton Mine, Copper Cliff, Ont. Considered to be outcrop of 23B$_1$

24B$_1$ Trap rock, Lat. 44° 56½′ N., Long. 109° 29½′ W., elev. 9800 feet, collected from a small working quarry by A. C. Lane, July 1937, 2 feet from the margin of a 100-foot dike at Long Lake, Wyoming. The dikes of this series are intrusive into the early pre-Cambrian complex (Stillwater series) and appear to be older than the middle Cambrian rocks of Beartooth Butte, but the exact relations are obliterated by overburden for a considerable distance. Age: possibly pre-Middle Cambrian, and post early pre-Cambrian.

24B$_2$ Trap rock, ditto 24B$_1$ except taken from the center of the 100-foot dike.

25B Trap rock, Lat. 44° 58′ N., Long. 109° 29′ W., elev. 10,600 feet, collected by A. C. Lane, July 1937, as part of Beartooth Range suite, from center of 25-foot dike, dip 45°, strike NNW.

26B Monchiquite dike, Lat. 55° 45′ N., Long. 5° 3′ W., submitted by Arthur Holmes in 1936 from Riasg Buidhe on the Island of Colonsay, Scotland. Geologic relations nowhere provide evidence of age beyond the fact that it cuts formations regarded as Torridonian. Age: Post-Devonian.

27B Monchiquite dike, ditto 26B but from Kilchattan instead of Riasg Buidhe.

28B Gabbro-diorite, 50-pound specimen collected by C. Goodman from General Crush Stone Quarry, Woburn, Mass. One of standard samples; *see* 3A.

29B Sudbury gabbro collected by H. C. Cooke, 1939, from 10 feet below original surface in road cut, lot 3, concession III, Graham Township, Ont.

30B Dunite, 50-pound specimen collected through the courtesy of the U. S. Geological Survey, from near Balsam Gap, N. Carolina. A part of the Webster dunite body. Composed dominantly of high magnesium olivine; a few per cent of chlorite, vermiculite, and chromite are present. Age is in question; some hold it to be pre-Cambrian; others that it is probably Ordovician. One of the standard samples; *see* 3A.

31B Kimberlite, 50-pound specimen obtained through the courtesy of U. S. Geological Survey and S. J. Shand, from Kimberly, S. Africa. One of standard samples; *see* 3A. Fragmental rock composed of olivine, its alteration products consisting largely of serpentine. The only important primary mineral, other than olivine, is a very pale-colored type of phlogopite. Fragments of several invaded rocks are also present. Some of the olivine is completely altered, but other grains up to several mm. in diameter are largely fresh with only veinlets of serpentine. Age: lower Cretaceous.

32B Andesite collected by K. G. Bell (1939, p. 10) from Dalhousie Mtn. in Nova Scotia.

33B Andesite of unknown origin.

34B Camptonite dike collected by R. J. Lougee from quarry in Burlington, Vt., visited by A-1 excursion of 1933 International Geological Congress. Cuts Cambrian quartzite; age possibly Triassic.

35B Diorite collected by G. D. Louderback from Wutchumna Hill, north side, Lindsay-Strathmore canal, ¾ mile NE of Lemon Grove, Calif., Lemon Grove quadrangle. Holocrystalline, granitic texture, medium grain, hornblende-hypersthene diorite. For petrographic and other radioactivity measurements *see* Finney (1934, p. 49).

36B Pumiceous lava with sparse phenocrysts of olivine belonging to the flow from Halemaumau of March, 1921, submitted as a powdered specimen by C. S. Piggot for intercheck measurements; Piggot's mean value, 0.94×10^{-12} g Ra/g, and geologic notes previously reported (Piggot, 1931, p. 3) using designation P-10.

37B Tower Falls basalt collected by A. C. Lane, 1937, in Yellowstone Park at about 1.4 of Guide Book (Bighorn Basin Yellowstone Valley Tectonics Field Conference, Aug. 3-5, 1937), 134 feet vertically and 30 feet horizontally from the outcrop surface. Age: Recent.

38B Trap, Lat. 44° 56½′ N., Long. 109° 29½′ W., elev. 9800 feet, collected by A. C. Lane, as part of Beartooth Range suite, from altered glassy margin of Long Lake Dike (*see also* 24B$_1$) by new road from Red Lodge to Yellowstone, near Long Lake, Montana, at about 35 of Guide Book (*see* 37B). Age: Possibly latest pre-Cambrian.

39B Trap rock from Gogebic range sill, Mikado Mine, Wakefield, Mich. From drill hole No. C-18 at 105 to 110 feet below collar; hole vertical and showing about 20 feet of drift above ledge. Location: SE-NW, Sec. 18-47-45, Mich. Specimen submitted by A. C. Lane.

40B Trap rock from Gogebic range sill, Mikado Mine, Wakefield, Mich. From drill hole No. G-48 at 855 to 860 feet below collar; hole vertical and showing about 20 feet of drift above ledge. This specimen is a more acidic phase than 39B. Location: NE corner of SE-NW, Sec. 18-47-45, Mich. Specimen submitted by A. C. Lane.

41B Trap rock from Gogebic range, Mich., collected by L. M. Scofield, 1936. *See* Lane (1938, p. 66) for additional geologic notes and other radioactivity measurements. Lane's designation: No. 31.

42B Trap rock from Gogebic range, Mich., same suite as 41B. Lane's designation: No. 27.

43B Trap rock from Gogebic range, Mich., collected by L. M. Scofield, Jan. 16, 1936. "Normal" from the 26th level of the Brotherton mine, cuts the upper phase of the Palms (Huronian). Lane's designation: No. 37; *see* 41B and 42B.

Igneous Rocks Intermediate Between Acidic and Basic

1I Trachyte collected by A. D. Howard from SE base of Cresent Hill, Yellowstone Park.

2I Dacite porphyry collected by A. D. Howard from summit of Bunsen Park, Yellowstone Park.

3I Gray tuff collected by W. D. Urry and W. L. Whitehead, Oct, 1935, 1 foot below glaciated surface and 2 feet above fossiliferous shale bed at fresh road cut, Newburyport Turnpike and Crowley Street, Rowley, Mass. Age: lower Devonian or upper Silurian, provisionally placed on dividing line.

4I Granodiorite from La Motte, Quebec collection of R. A. Daly. Other measurements by Keevil (1938, p. 145).

5I Tholeiite from Cleveland Dyke, Bolam County, Durham, England, submitted as a small hand specimen for intercheck measurements by Arthur Holmes (1937, p. 118); *see* 4B.

6I Hornblende granodiorite, Lat. 48° 14′ N., Long. 77° 54′ W., collected by W. C. Gussow in 1936 from a specimen freshly blasted from a road cut 10 feet from the surface. The granodiorite is intrusive into a sedimentary complex of Keewatin or possibly earlier age and is thought to be older than the diabase dikes in the area and the Cobalt series to the southwest. Age: Probably pre-Cobalt and post-Keewatin.

7I Quartz dolerite from Whin Sill, Scordale Beck, Westmoreland, England, submitted as a small hand specimen for intercheck measurements by Arthur Holmes (1937, p. 118); *see* 4B.

Metamorphic Rocks

1M Gneiss from Taroko-kyo, collection of Z. Hatuda; *see* 16A.

2M Gneiss collected by T. F. W. Barth in Finland, designated as "augen Gneis" and classified as a migmatite. Lead has been quantitatively separated from a large sample of this rock by E. Gleditsch and coworkers and submitted to A. O. Nier for isotopic analysis in an attempt to determine the age by the lead method.

3M Eclogite from Saastal, Switzerland, submitted as a small hand specimen for intercheck radioactivity measurements by J. H. J. Poole, who obtained 0.42×10^{-12} g Ra/g (personal communication, 1938); *see* 5A.

SEDIMENTARY ROCKS

1S Limestone submitted as a powdered specimen by F. Behounek, Director of State Radiological Institute of Czechoslovakia in 1936 for intercheck measurements. Behounek (personal communication, 1937) reports 24×10^{-12} g Ra/g and designates as "Silurian Bituminous Limestone". We observed the decrease in weight upon heating at 1000° C in air for 1 hour was 4.0 and 5.5 per cent for two portions of specimen.

$2S_1$ Shaly limestone core from 3780-00 feet, Moore No. 7 Schauers, Fitts Pool, Oklahoma. Age: Ordovician.

$2S_2$ Shaly limestone core from 3810-35 feet, Moore 1A Harper, Fitts Pool, Oklahoma. Age: Ordovician.

$3S_1$ Limestone, shale, quartz core from 4265-70 feet, Moore No. 7 Schauers, Fitts Pool, Oklahoma. Age: Ordovician.

$3S_2$ Shaly limestone, sandstone core from 4245-81 feet, Moore 1A Harper, Fitts Pool, Oklahoma. Age: Ordovician.

4S Dolomite core from 4370-75 feet, Moore No. 7 Schauers, Fitts Pool, Oklahoma. Age: Ordovician.

5S Carthage Limestone, 50-pound specimen obtained through the courtesy of the U. S. Geological Survey from Carthage, Missouri. One of standard samples, see 3A. Composed exclusively of calcite with obvious fossil fragments of which crinoid remains are dominant. So completely cemented that no pore space is visible under the microscope; grains about 0.5 mm.

$6S_1$ Quartz sandstone core from 5108-16 feet, McKinney No. 5, Sun Field, Starr Co., Texas. Age: Oligocene.

$6S_2$ Quartz sandstone core from 4645-50 feet, Olivares No. 5, Sun Field, Starr Co., Texas. Age: Oligocene.

$6S_3$ Quartz sandstone core from 4657-8 feet, McKinney No. 6, Sun Field, Starr Co., Texas. Age: Oligocene.

$7S_1$ Sandstone core from 494 feet, Nowata Co., Oklahoma. Age: Pennsylvanian.

$7S_2$ Ditto $7S_1$ from 505 feet.

$7S_3$ Ditto $7S_1$ from 512 feet.

8S Shaly sandstone core from 2510-40 feet, Moore No. 7 Schauers, Fitts Pool, Oklahoma. Age: Pennsylvanian.

9S Berea sandstone, 50-pound specimen obtained through the courtesy of the U. S. Geological Survey from Cleveland, Ohio. One of standard samples; *see* 3A. Composed of subangular sand grains, a little chertlike quartz, and less calcite and feldspar. Age: Mississippian.

$10S_1$ Quartz sandstone core from 5889-94 feet, No. 1 Dailey, Navarro Crossing, Texas. Age: Cretaceous.

$10S_2$ Quartz standstone core from 5934-35 feet, No. 1 C. C. Hill, Navarro Crossing, Texas. Age: Cretaceous.

11S Quartzite, 50-pound specimen obtained through the courtesy of the U. S. Geological Survey from Bull Run Mountain, Va. One of standard samples; *see* 3A. Composed of quartz in interlocking grains (thoroughly recrystallized), a little muscovite in parallel arrangement, and a few grains of magnetite.

$12S_1$ Clay shale core from 5096-7 feet, McKinney No. 5, Sun Field, Starr Co., Texas. Age: Oligocene.

$12S_2$ Clay shale core from 4649-53 feet, McKinney No. 6, Sun Field, Starr Co., Texas. Age: Oligocene.

$12S_3$ Clay shale core from 4636-7 feet, Olivares No. 5, Sun Field, Starr Co., Texas. Age: Oligocene.

$13S_1$ Shale core from 5851-67 feet, No. 1 Dailey, Navarro Crossing, Texas. Age: Cretaceous.

$13S_2$ Shale core from 5916-17 feet, No. 1 C. C. Hill, Navarro Crossing, Texas. Age: Cretaceous.

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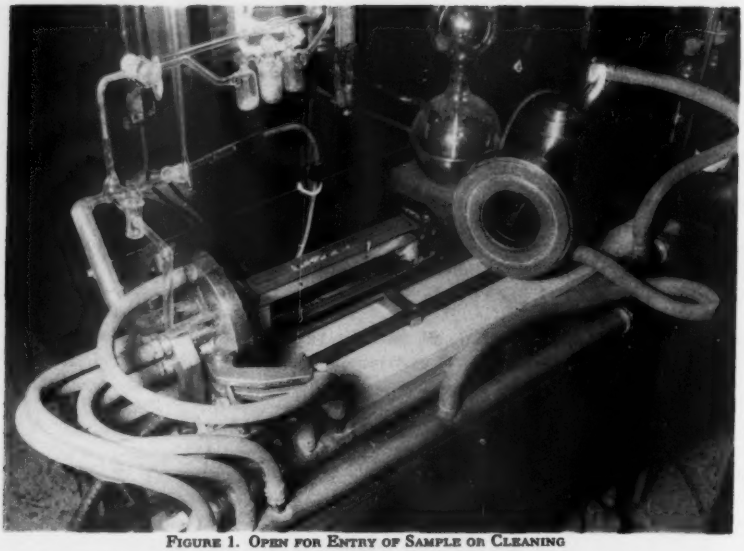

FIGURE 1. OPEN FOR ENTRY OF SAMPLE OR CLEANING

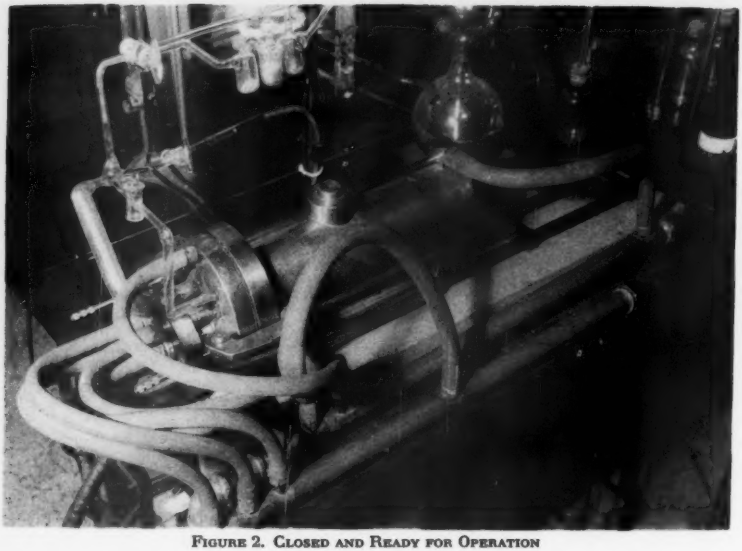

FIGURE 2. CLOSED AND READY FOR OPERATION

DIRECT-FUSION, VACUUM FURNACE

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AGE MEASUREMENTS BY RADIOACTIVITY

BY CLARK GOODMAN AND ROBLEY D. EVANS

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ABSTRACT

The radioactive methods of geologic age determination involve the four basic requirements: (1) a known, systematic rate of disintegration of the radioactive elements; (2) accurate measurement and sampling; (3) absence of disintegration product as a primary constituent; (4) no addition or subtraction of the disintegration product or its source during the history of the material. A critical review is presented of our knowledge of the fulfillment of these requirements by the various radioactive methods. Emphasis is placed on the helium method because of its possible wide applicability, because no adequate review has heretofore appeared, and because considerable confusion exists concerning the present status of this method.

Following a discussion of the necessity for completely discarding all of Urry's helium age measurements, a summary of the remaining data shows that (1) with only a few exceptions the helium age ratios lie in the proper geologic sequence when the results for acidic rocks and for basic rocks are considered separately, (2) the age ratios for acidic rocks are consistently lower than the age ratios for corresponding basic rocks, and (3) the helium age ratios, even for basic rocks, are substantially lower than corresponding lead ages on radioactive minerals. These general inconsistencies stimulated the present reinvestigation of fundamental requirements underlying the radioactive methods. The retentivity of rocks for helium has been found to be the major source of uncertainty in the present application of this method. A review of the lead method indicates that only comparatively few ages can be considered as possible fixed points in the lead time scale. The uncertainty in the isotopic constitution of ordinary lead is seldom an important source of error, but the need for isotopic analysis in all lead age measurements is emphasized by the relative insensitivity of atomic weight determinations. Further work needs to be done on the effects of leaching and weathering of radioactive minerals. Brief reviews of the isotopic ratio, rubidium-strontium, and possible potassium methods of age determination are included for the sake of completeness.

ACKNOWLEDGMENTS

A number of individuals have aided in the measurements reported and the clarification of ideas expressed in this paper. The writers are indebted to Dr. P. M. Hurley and Mr. C. R. Deibert for their analytical aid. The generous criticism of Professors Adolph Knopf and J. C. Boyce and Dr. J. P. Marble in reviewing the original manuscript and the interest and encouragement shown by Professors W. J. Mead and A. C. Lane is appreciated. This research has been supported in part by the Geological Society of America through grants-in-aid to Professors A. C. Lane and W. J. Mead.

INTRODUCTION

By the united application of the three main subdivisions of Historical Geology—Stratigraphy, Lithology, and Structural Geology—the relative sequence of many of the geological formations of the lithosphere have been established.

The evidence from Paleontology, strictly speaking a branch of Stratigraphy, is so distinctive that our extensive post-Proterozoic time scale is based on the characteristic forms of life which existed during each geologic period. However, the igneous rocks, which constitute more than 95 per cent of the earth's crust, contain no fossils and can be accurately placed in the geologic time scale only if there is a clear-cut structural relation to fossil-bearing sedimentary rocks. The vast expanse of pre-Cambrian time is represented by practically no fossils.

The structural and paleontological methods, however, are inherently unable to give more than a qualitative indication of age—that is, of elapsed time in thousands or millions of years. A number of important results accrue from the establishment of a quantitative time scale. It provides a measure of the time required for the biological changes observed in the study of paleontology. A knowledge of the length of time involved in decomposition, disintegration, and deposition is of value in the study of Sedimentation. Similarly, the quantitative knowledge of ages serves to measure the rate of diastrophic processes, which is of interest to the structural geologist and petrologist. A more exact span can be assigned to certain of the periods and epochs than can be obtained from paleontological evidence, and a truer estimate of their relative lengths thus obtained. But the most valuable contribution to geology will come from the application of the quantitative method to the host of rocks that cannot otherwise be dated or even correlated. This need is especially acute for pre-Cambrian rocks.

The systematic disintegration of the geologically long-lived radioactive elements provides the most reliable method of measuring geologic time. Although we shall consider briefly methods based on the disintegration of some of the lighter elements, our discussion will mainly concern itself with age measurements based on the disintegration of the two parent elements, uranium and thorium, and their daughter elements, represented schematically as shown in Figure 1.

It is seen that the element uranium is composed of two main isotopes,[1] uranium I (UI) and actino-uranium (AcU), with atomic weights of 238 and 235 respectively. The most accurate measurement of the relative

[1] Isotopes of a given element have the same atomic number, hence possess the same chemical properties, but have different atomic weights (indicated by superscripts).

abundance of these two isotopes (Nier, 1939, p. 150) indicates 99.29 per cent UI and 0.71 per cent AcU. The transmutation of one atom of UI, through the uranium series, into a stable lead isotope, Pb^{206}, produces as a by-product 8 atoms of helium, He. Similarly, an atom of AcU

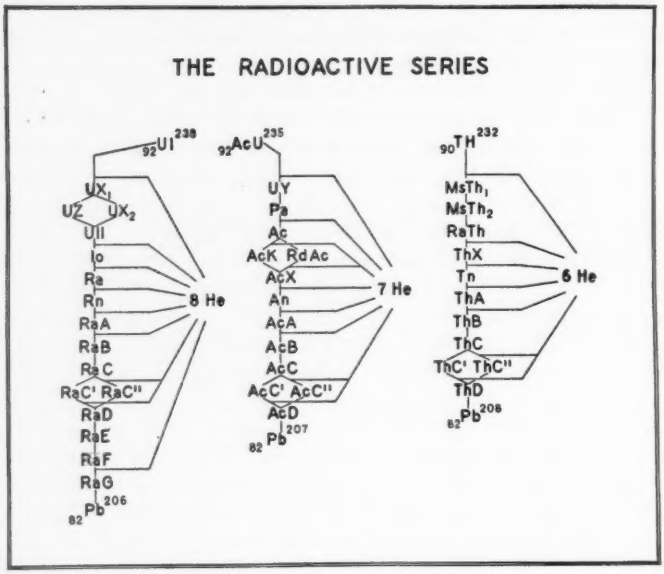


FIGURE 1.—*The radioactive series*

ends as Pb^{207} and produces 7 He, and an atom of Th ends as Pb^{208} and produces 6 He.

This systematic generation of lead isotope end-products and of helium as a by-product are the properties of these parent radioactive elements which make possible their use as indicators of geologic time. The age equation can be expressed in the fundamental form:

$$Age = \frac{Amount\ of\ disintegration\ product}{Rate\ of\ production\ of\ product} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

in which the numerator represents the amount of radiogenic lead or helium formed and the denominator measures the rate, expressed in the proper units, at which the disintegration product is being generated in the material whose age is being measured. The *Age* refers to the time that has elapsed since final solidification or crystallization of the rock or mineral containing the radioactive elements. The application of this

equation to terrestrial materials involves the four basic requirements: (1) a known, systematic rate of disintegration of the radioactive elements; (2) accurate measurement and sampling; (3) absence of disintegration product as a primary constituent; (4) no addition or subtraction of the disintegration product or its source during the history of the material.

In the light of present-day knowledge of nuclear physics, the first requirement is clearly well established. All efforts to alter the rate of radioactive decay have invariably led to negative results from which it is concluded that radioactive decay proceeds at the same rate independent of physical environment within the earth. Evidence from pleochroic haloes proves that the rates of decay of radioactive elements have been constant over geological periods of time.

Evaluation of the extent to which the other three requirements are fulfilled by different geological materials has been the subject of considerable research and constitutes the major discussion of this paper. The problem divides itself into two parts: (1) the helium method as applied to ordinary rocks and minerals, and (2) the lead method as applied to radioactive minerals.[2] The reason for this general division of applicability can be understood from Figure 2. On a vertical logarithmic scale, representative total uranium (U)—*i. e.*, UI + AcU—and thorium (Th) contents for radioactive minerals, igneous rocks,[3] and a hypothetical rock, to be discussed later, are indicated in each case by the two solid lines on the left. The amounts of radiogenic lead (Pb*) and helium (He) that would be generated in these materials in 100 million years and the amounts of original ordinary lead (Pb) to be expected are indicated on the right. The lead content of such a radioactive mineral can quite readily be separated quantitatively and weighed by the usual chemical techniques. However, the minute lead content found in ordinary rocks is considerably beyond the range of chemical methods, as indicated by the schematic arrangement of Figure 2. Similarly the uranium and thorium content of radioactive minerals can be determined gravimetrically, but the content of these elements in ordinary rocks can be determined only by physical methods using sensitive electrical detection apparatus.

The helium content, corresponding to chemically separable quantities of radiogenic lead, would be several cc per gram.[4] Hence, it is hardly surprising that an appreciable fraction of the helium generated in even relatively young radioactive minerals has escaped, since the internal

[2] Throughout this discussion the term "radioactive mineral" is intended to designate terrestrial materials that contain uranium or thorium as important constituents, present to the extent of 0.01 per cent or more, as distinct from rocks or minerals that contain only traces of these elements.

[3] The U and Th values for the igneous rock lie midway between the averages for acidic and basic rocks given by Evans and Goodman (1941a, Table 10).

[4] In age measurements, helium volumes are referred to 0° C. and 760 mm. of Hg pressure.

pressure often reaches several atmospheres. It is this widely different nature of the two decay products, lead and helium, that divides the applicability of the two methods of geological age determination. Thus, in

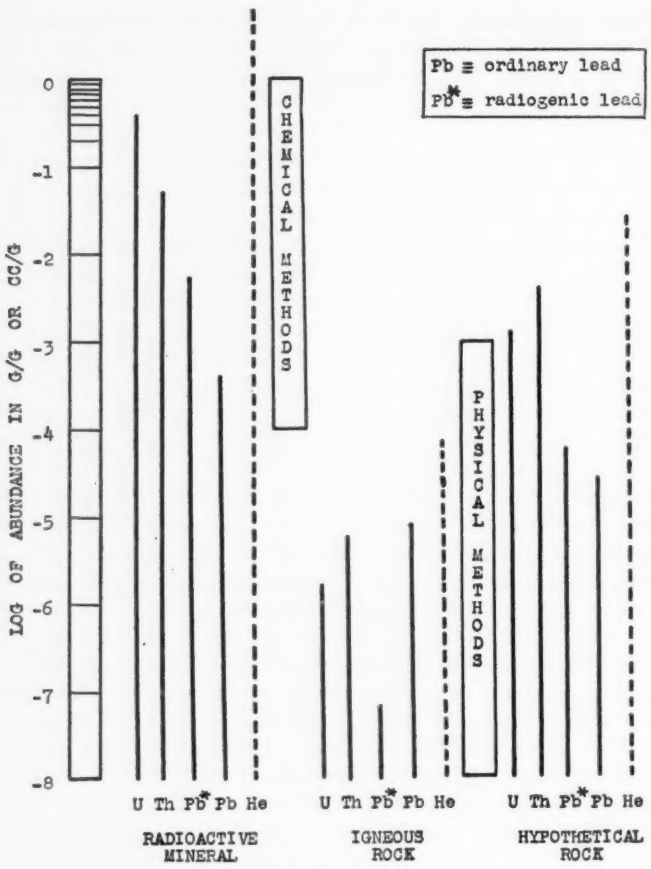


FIGURE 2.—*Schematic representation of radioactive content and decay products*

general, the lead method is limited to radioactive minerals, and the helium method to ordinary rocks and minerals. In the following sections, an attempt is made to evaluate the extent to which the fundamental requirements are fulfilled by the two methods.

By fortuitous circumstances a material might be found with an intermediate composition, such as that shown for the hypothetical rock in Fig. 2. In this case both the helium and lead methods might be applicable to the same specimen. Thus far a direct comparison of this type has not been made.

HELIUM METHOD

GENERAL CONSIDERATIONS

Since the discovery by Strutt (1908, p. 272) that the helium content of radioactive minerals could be correlated with their probable geological age, considerable work has been devoted to the application of this principle to the measurement of geologic time. It was shown quite early (Strutt, 1909, p. 166) that some helium escapes from radioactive minerals, even at room temperature and atmospheric pressure. This loss of helium lowers the numerator of our age equation and, hence, the resulting ratio is lower than the true age of the radioactive mineral. For this reason, age ratios on such minerals usually only indicate minimal values for the age.[5] As we have seen from Figure 2 and have mentioned in the introduction, the internal pressure of helium associated with these high concentrations of uranium and thorium is so great that there is little wonder some helium escapes. Rather, the large quantities retained under these conditions over geologic time attest to the inherently high retention of helium in these minerals. The early investigators did not appreciate this viewpoint and did not have sufficiently sensitive apparatus to extend the helium method to igneous rocks. As a consequence, the helium method lay in disrepute for nearly 20 years following Strutt's classical work.

While radioactive minerals undoubtedly do not fulfill the fourth fundamental requirement concerning helium, it does not necessarily follow that the same is true of igneous rocks, where the concentrations of helium, uranium, and thorium, as we have seen, are only about one-millionth of those found in radioactive minerals. With this in mind, Dubey and Holmes (1929, p. 794) made the first helium age determinations on igneous rocks. The results were so encouraging that a re-exploration of the long neglected helium method was begun. During the interval following Strutt's initial researches, the analytical techniques required for measuring the minute quantities of helium and radioactivity in rocks had been developed. Paneth and his coworkers (*see* Paneth and Urry,

[5] Throughout this paper and the one by Hurley and Goodman (1941), we are faced with the problem of differentiating between the age indicated by experimental measurements and the true numerical age of the geological material, which, we shall see, may be quite different. Accordingly, the term, age, represents the true age and the term, age ratio, the calculated quantity. If the four fundamental requirements are fulfilled for a given material, the age and the age ratio will, of course, be equal.

1930, p. 110, for references) had greatly improved the technique for the isolation and measurement of small quantities of helium. Joly, Poole, Piggot, Paneth, and Evans had developed methods of measuring the minute radioactivities involved. Thus, rapid strides in the application of the helium method were possible because of the availability of the necessary detection instruments.

Under the guidance of the National Research Council Committee on the Determination of Geologic Time by Atomic Disintegration, A. C. Lane, Chairman, and supported by the American Philosophical Society and the Geological Society of America, an intensive program of helium age measurements was begun. Between 1932 and 1936 an elaborate helium time scale evolved largely as the result of measurements by W. D. Urry. (*See* Urry, 1936, p. 1219, for references.) Some investigations of the basic geological assumptions underlying the helium method were made during this time. In general, however, the establishment of the validity of the method rested upon the accumulation of a large number of apparently concordant measurements. The most convincing evidence of reliability came from the apparent agreement with corresponding ages based on lead ratios in radioactive minerals. Thus it appeared that a reliable time scale had been established which could be used as a reference for all geological age measurements.

In the spring of 1936 the researches herein reported were begun. The direct-fusion furnace, which had proved so satisfactory in the liberation of radon from rocks (Evans, 1935, p. 102) appeared to offer similar advantages in the determination of helium. Preliminary alpha-activity measurements on rocks (Evans, 1934 a and b, p. 29-42; Finney, 1934, p. 1-98; Finney and Evans, 1935, p. 503-511; Raitt, 1935, p. 1-78) suggested the feasibility of this method of determining the total rate of helium production—*i. e.*, the denominator of the age equation. The combination of these two techniques offered a direct, physical method of age determinations which would be independent of radioactive standards and much more rapid than the radon-thoron-helium method used by Urry (1936, p. 34-48). Since mass production of age results seemed in order, the development of assembly-line methods appeared appropriate.

Initial measurements by the alpha-helium method, on rocks previously analyzed by Urry, gave age ratios lower by a factor of about two. Further study indicated that this difference was primarily due to the larger activity index [6] obtained by the alpha-counting method as compared with that resulting from Urry's radium and thorium measurements. At this

[6] In the alpha-helium method the rate of production of helium is observed as the number of alpha particles (helium nuclei) emitted per hour per milligram of rock, which we define as the "activity index." This quantity is determined indirectly in the radon-thoron-helium method from the measured radon and thoron contents.

time the most likely explanation for the differences appeared to be the presence of a previously undiscovered radioactive series in terrestrial materials. This exciting possibility stimulated an investigation into the exact nature of the analytical differences. An extensive program of calibration and intercomparison of measurements on the same rock samples was carried out between the two methods. The results of this investigation have already been published by Evans, Goodman, Keevil, Lane, and Urry (1939, p. 931-946). It was established that there are no unknown strong alpha-emitting radioactive elements in ordinary terrestrial materials.[7] This conclusion follows from the good agreement between activity-index measurements made by direct alpha counting and those based on separate radon and thoron measurements of the same rock samples. This agreement appeared only after Urry's radium apparatus had been recalibrated, the earlier calibrations having been found to be in error by a factor of over two. As previously mentioned (Evans and Goodman, 1941a, p. 470-471) part of this discrepancy can be explained by the use of unreliable radium standards, but the remaining difference has not yet been resolved.

With a constant Th/U ratio, the use of a constant radium correction factor would lower evenly all the earlier helium ages. However, as is evident from the Th/U values given by Evans and Goodman (1941a, Tables 6-9), this ratio varies considerably, and a correction, applied to the radium measurements alone, would cause an uneven decrease in Urry's helium time scale. Another difficulty is that when the earlier radium measurements are recomputed, using the corrected calibration constant, the ages based on these values are also higher than more recent measurements (by both the alpha-helium and radon-thoron-helium methods) on the same hand samples (Evans *et al.*, 1939, Table XI, p. 943).

The apparent agreement between ages based on the lead method as applied to radioactive minerals and Urry's helium ages on fine-grained igneous rocks was cited (Urry, 1936, p. 1230; Holmes, 1937, p. 179) as the most cogent evidence for the validity of both methods. With the downward revision of helium age ratios necessitated by the newer studies, this apparent agreement, which we now know was merely fortuitous, disappeared. In its stead we were left with only a limited number of reliable values constituting a series of helium age ratios that appeared to be in a sequence consistent with geologic evidence, but in magnitude at variance with the corresponding lead ages. At this juncture two possibilities appeared likely—either the lead method involved some previously undisclosed error, or one or more of the fundamental requirements

[7] Evans and Raitt (1935, p. 174) have previously shown that there are no strong unknown gamma-ray emitters in terrestrial materials.

of the helium method had not been fulfilled by the rock samples measured. An inquiry into the lead method, most of which is included in a later section of this paper, showed that while considerable uncertainty exists in many of the lead age ratios, some are unusually well authenticated. Attention was then turned to a critical evaluation of the various uncertainties underlying the helium method, a summary of which was presented before the Geological Society (Mead, Evans, and Goodman, 1939, p. 1921), and the details of which are given in subsequent sections of the present paper. In brief, it was concluded that with the proper care the second requirement of accurate measurement and sampling can be fulfilled. The possible presence of primary helium can be avoided by a not too restrictive selection of material, thus satisfying the third requirement. The fourth requirement, particularly as it pertains to the loss of helium, remained as the major source of uncertainty in the application of the helium method to rocks. Accordingly, a systematic program of research was undertaken, in collaboration with Dr. P. M. Hurley, which was specifically designed to test the fulfillment of the fourth requirement by various rocks and minerals. The details of this work are presented by Hurley and Goodman (1941, p. 549-556) but, for the sake of completeness in the present consideration, are also summarized here as follows:

The retentivity of rocks for helium has been found to be extremely variable and is the major source of uncertainty in the application of this method. Researches on separate mineral components indicate that, with only a few exceptions, helium ages obtained on rocks as a whole, even fine-grained, unaltered mafic rocks, can represent only minimal values. Certain minerals, notably the feldspars, appear to lose a large fraction of their radiogenic helium. On the other hand, certain of the mafic minerals, pyroxene and magnetite in particular, have a much higher retentivity and show helium ages which approach the values to be expected from the lead time scale. Thus, all helium ages made on rocks as a whole represent mean values dependent upon the proportion and helium retentivity of the mineral components. It is anticipated that further research on selected rock minerals will yield helium ages suitable for the establishment of a new helium time scale in substantial agreement with the present lead time scale.

DETERMINATION OF HELIUM

General statement.—We have seen that the fundamental age equation involves knowledge of the helium content, in the numerator, and the rate of production of helium, in the denominator. In this section the techniques employed for the measurement of the helium will be briefly reviewed.

Most rocks contain between 10^{-6} and 10^{-4} cc of helium per gram. The determination of these minute quantities involves: (1) liberation, (2) purification, and (3) measurement. There are a number of ways in which

these three steps can be performed, but the techniques summarized below appear to offer the optimum combination of reliability and convenience.

Liberation of gases.—The development of the direct-fusion furnace and its application to radioactivity measurements have been described previously by Evans (1935, p. 102). In principle the same type of furnace is used in the release of helium from terrestrial materials. However, because of the significant helium content of the atmosphere (*see* footnote 16), a vacuum-tight design is necessary. For this reason, the earlier methods of sealing have been superseded by a double-gasketed face plate and special inset electrode seals. These and other features can be seen in the photographs (Pl. 1). In Figure 1 of Plate 1, the furnace is opened, and the upper graphite radiation screen removed to show the mounting of the graphite crucible, the glass vacuum leads, and the rubber hose water connections for cooling. The swinging bed allows convenient removal of the sublimate that lines the furnace after each fusion of rock. In Figure 2 of Plate 1, the furnace is closed and ready for action; the mercury bubbler trap is seen just above the peep-hole in the top of the furnace.

A large volume of gas, of which the helium constitutes only a minute fraction, is released during the fusion of a rock sample. This gas acts as a flushing medium in quantitatively sweeping all helium out of the furnace and through the mercury bubbler trap which prevents back diffusion.

Experimental evidence for the quantitative release of helium from rocks by direct fusion (at 1500° to 2000° C.) is afforded by the absence of helium in repeat measurements on the rock melt at elevated temperatures (2000° to 2500° C.). Studies of the fractional liberation as a function of temperature indicate that 99 per cent or more of the helium is released from most rocks within 5 minutes at temperatures only slightly above 1500° C. It is routine practice to heat the sample for at least 5 minutes at a temperature of 2000° C. or above. Comparative measurements on quartered portions of a number of granulated rock samples show good agreement between the carbonate-fusion and direct-fusion techniques (Evans, *et al.*, 1939, Table VIII, p. 941).

Purification.—Claude (1914, p. 861) was the first to conduct extensive research on the purification of the noble gases. He showed that helium and neon are absorbed to a negligible amount on charcoal at liquid air temperatures when compared with the other inert gases and hydrogen. Paneth and Urry (1930, p. 110) applied this method to the separation of pure helium from the other gases released when meteorites were dissolved in acid or fused with alkali carbonates.

The most troublesome contaminant is hydrogen, since it is only partially absorbed on the charcoal. The method previously used (Paneth

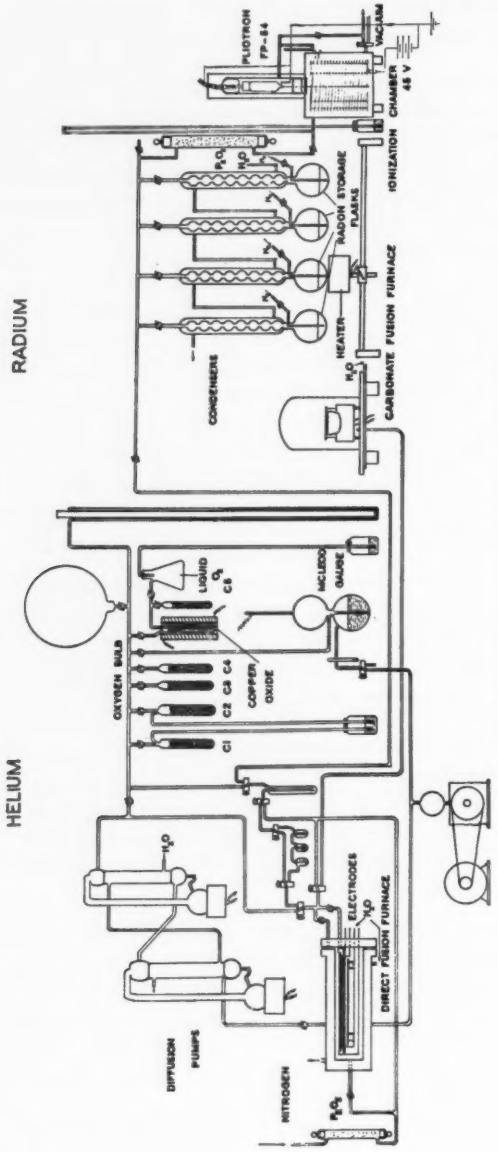


FIGURE 3.—*Diagram of helium and radium apparatus*

and Urry, 1930, p. 118; Urry, 1933, p. 340) for removing the last traces of hydrogen was the introduction of pure, electrolytic oxygen followed by circulation of the gases over heated palladium (approximately 300° C.). This procedure catalyzes the formation of water vapor which is subsequently absorbed on charcoal together with any excess oxygen.

A bulb containing liquid oxygen has been found to be a somewhat more convenient and equally pure source of oxygen. The general arrangement of the apparatus is represented by the left-hand side of Figure 3. The liquid oxygen is poured into a flash equipped with two outlet tubes, one leading to a bubbler through which the excess gas can escape, the other leading into the main body of the system through a pyrex tube containing stick cupric oxide. The cupric oxide, when heated, serves the same purpose as the palladium but is not susceptible to poisoning and hence does not decrease in efficiency with use.

The charcoal tubes (Fig. 3, C1-C5) dipped in liquid air are used in successive order for absorption of all the gases except helium, which remains distributed throughout the vacuum-tight, pyrex system.

Measurement.—After final purification the volume of helium is measured in the modified McLeod gauge equipped with a ground capillary (Rosenberg, 1938, p. 258). This type of capillary has been found to have a more uniform correction (for capillarity) and hence yields more reproducible results than the smooth-walled variety used by other workers. Ten or more measurements of the length of the gas column in the capillary are made at various pressures, observed by the differential height of the mercury in the side-arm tube. These observations, after the capillary correction is applied, usually show a maximum variation of less than 5 per cent from the mean value of the pressure-volume product. By the use of the perfect gas law, involving the observed temperature and calibration constant of the apparatus,[8] the volume of helium contained in the known weight of rock sample can be readily calculated.

An accurate estimate of the experimental uncertainty in helium measurements on rocks is difficult to make, largely because of the somewhat inhomogeneous distribution of helium in rocks. Repeat measurements on separate portions of the same hand specimen usually agree well within a probable error of 10 per cent of the mean, and on quartered portions of the same sample within 5 per cent of the mean. Some rocks, however, show variations in helium distribution of several times this amount. The relatively narrow range of values observed for the calibration constant (Evans, *et al.*, 1939, Table VII, p. 941) indicates that the

[8] The calibration constant is defined as the ratio of the effective total volume of the apparatus to the volume of the McLeod gauge. The capillary is calibrated by weighing measured lengths of mercury at a known temperature. The diameter of the bore generally used is about 0.25 mm.

purification procedure is quantitative to within 2 per cent for moderate volumes of helium, around 10^{-4} cc. Thus for most measurements the sampling error is predominant. For volumes of 10^{-6} cc of helium, the estimated probable error amounts to about 30 per cent.

DETERMINATION OF RADIOACTIVITY

Methods.—Two methods can be used to determine the rate of production of helium in terrestrial materials. The older method involves separate measurements of the uranium and thorium and the use of their decay constants to calculate the corresponding rate at which helium is being produced. In actual practice the radon and thoron contents are determined, and the proportion of the parent elements calculated, assuming radioactive equilibrium. Since modern detection instruments make it readily possible to count individual alpha rays, a direct, physical method of determining the rate at which helium is being produced in terrestrial materials has been developed. This newer method possesses the distinct advantages of being free from chemical manipulation and having only a second-order dependence upon standard radium or thorium solutions and upon the decay constants of uranium, thorium, and actino-uranium, which enter only in a small correction term for very old rocks. Both these methods have been used for the age analyses reported in this paper and in the paper by Hurley and Goodman (1941). Consequently, a brief review of the two methods is included.

Alpha-count method.—Since the only known natural source of helium is from the alpha particles emitted by terrestrial radioactive sources,[9] the true rate of production of helium is given directly by the total alpha emission within a rock. However, it has already been shown by Finney and Evans (1935, p. 503) that such measurements are complicated by the small range of alphas in these materials and the variation of range for alphas from the different radioactive elements. For example, the alpha rays of uranium I have a mean range of only about 14 micra in rock, while the normal alphas from thorium C′ penetrate only about 46 micra. Except for the rare, long-range alphas from the C′ bodies and the very short range ones from samarium,[10] the alphas from the 20 other emitters lie between these limits. For sources thick to these alpha rays, the conversion of the observed count to the true emission depends directly upon an accurate knowledge of the stopping power of the rock, the proportion and ranges of the various alpha emitters, and an experimental

[9] The recently discovered (Alvarez and Cornog, 1939, p. 613) stable isotope, He^3, in ordinary helium has a relative abundance of less than 10^{-7}, and its origin is not yet known.

[10] In practically all rocks, the helium contributed by samarium is negligible compared with that from the other radioactive elements.

factor involving the minimum detectable range of alpha rays in the counting apparatus (Finney and Evans, 1935, p. 504). While each of these quantities is either known or can be measured within a probable

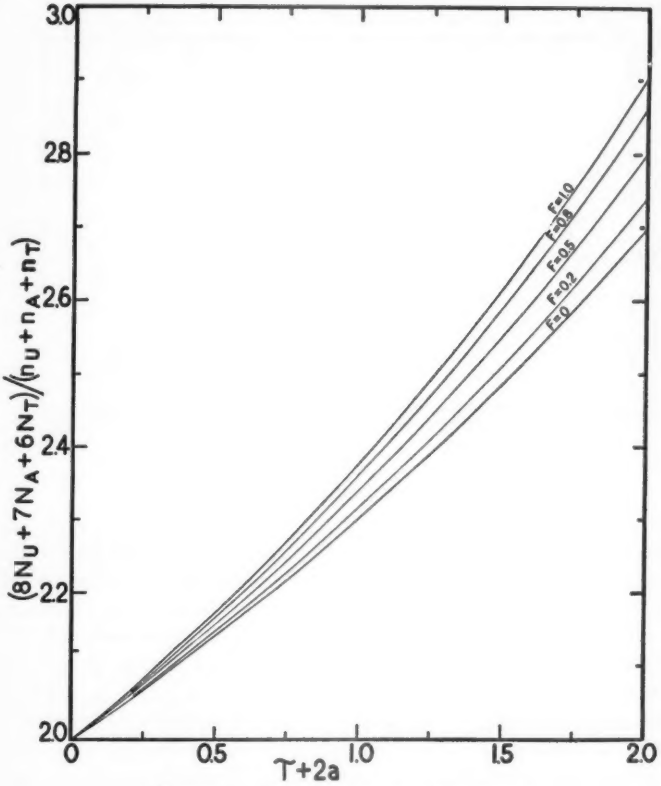


FIGURE 4.—*Alpha-count conversion curves*

error of about 10 per cent, the combined application results in considerable uncertainty of measurement for thick sources, which usually offsets the advantages to be derived from the alpha-counting method. Although rock sources sufficiently thin to cause only a fractional absorption of even the shortest range alphas give a somewhat lower counting rate than can be obtained from thick sources, the conversion of the observed count to the true emission only involves the above factors in terms of second-order importance. Consequently, considerable improvement in

accuracy is obtained by the use of thin rock sources, and the present discussion is limited exclusively to such deposits.

Following the general equations derived by Finney and Evans (1935, p. 503-511) which relate the observed counting rate to the true rate of

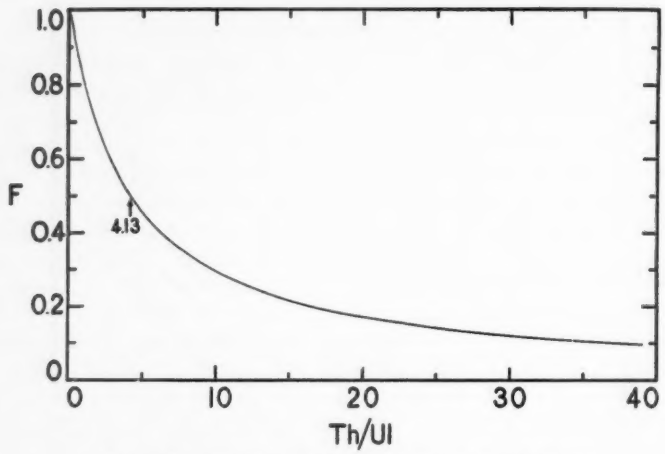


FIGURE 5.—*Graphical relationship between F and Th/UI*

emission of alpha particles within the source, the graphical relationships shown in Figure 4 can be established.[11] The ratio of the true to the observed rate of alpha emission, $(8N_U + 7N_A + 6N_T)/(n_U + n_A + n_T)$, is obtained for a given thickness of source τ plus absorber a, by interpolation between the curves for $F = 0$ to $F = 1.0$. The quantity F represents that fraction of the total alpha rays produced in the source due to the uranium and actino-uranium series. Consequently, values of F from 0 to 1.0 cover the range of all possible values and are related to the thorium-uranium ratio, Th/UI, as shown in Figure 5. At a Th/UI ratio[12] of 4.13—that is, $F = 0.5$—the rate of production of helium by the thorium series is equal to the rate of production of helium by the uranium plus actino-uranium series.

Referring again to Figure 4, it is seen that for a thickness of rock source plus absorber of $\tau + 2a = 1.0$ air-cm., which corresponds to slightly more than 5 micra, a maximum uncertainty of less than 2 per cent is

[11] The details of this derivation and a general description of the alpha-helium technique are being published in physical journals (Evans and Goodman, 1941b).

[12] Average values of 5.0 and 4.0 were obtained for the Th/U ratio (essentially the same as the Th/UI ratio) of acidic and basic rocks respectively by Evans and Goodman (1941a, p. 474).

introduced by assuming F = 0.5. Hence, for most rocks, accurate knowledge of the proportion of Th and U is unnecessary in determining the denominator of the age equation by direct alpha counting.

To obtain the approximate age ratio, t_o, in billions of years (10^9) from

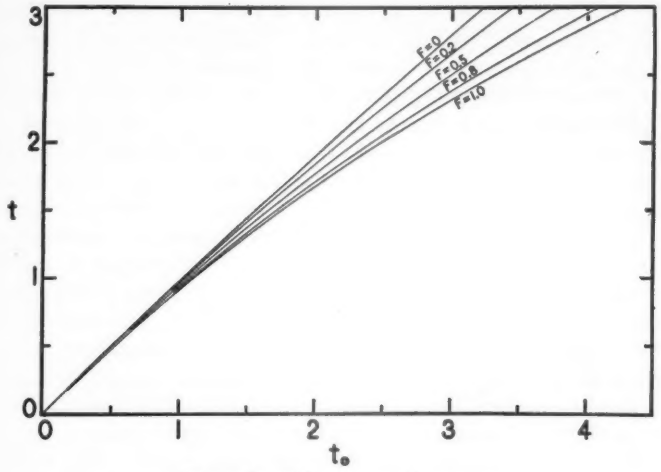


FIGURE 6.—*Old age correction curves*

the observed helium content, He, and activity index,[13] I, the general expression, Equation (1) is rewritten as follows:

$$Age = t_o = 0.0308 \frac{Helium\ in\ 10^{-5}cc/g\ of\ rock}{Activity\ Index\ in\ \alpha/hr \cdot mg}$$

$$t_o = 0.0308 \frac{He}{I} \qquad (2)$$

In all rocks the present-day rate of production of helium is less than in the past due to the wearing out of the very slowly decaying parent elements. This effect is not significant for rocks younger than about 500 million years but introduces an increasingly important correction as the age increases above this value. Thanks to the reliable researches of Kovarik and Adams and of Nier, we now have an accurate knowledge of the decay constants of uranium, thorium, and actino-uranium, which allows this old-age correction to be made with considerable certainty. The true age ratio, t, is obtained from the approximate age ratio, t_o, by reference to Figure 6. It is seen that, for ages $\lesssim$ 1 billion years (t = 1),

[13] In the alpha-helium method, I = (8Nu + 7Na + 6Nt) and is obtained by multiplying the observed rate of alpha emission, (n_s + n_a + n_t), by the conversion factor obtained from Fig. 5. In the radon-thoron-helium method, I = (1.04 Ra + 0.0886 Th) where Ra = radium content in 10^{-12}g/g of rock and Th = thorium content in 10^{-6}g/g of rock.

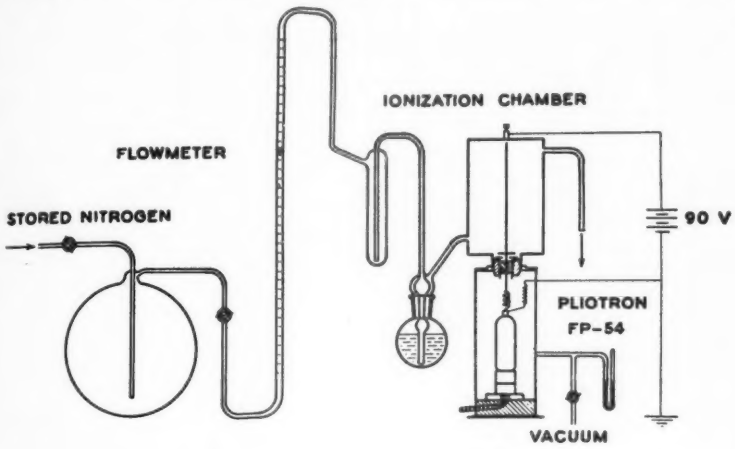
a maximum error of 5 per cent results from the assumption that $F = 0.5$. For greater ages, an accurate value of F can always be obtained from a radium determination, because: $F = \frac{1.04Ra}{I}$.

Radon-thoron method.—The direct-fusion technique of measuring the radium, and hence the uranium, content of rocks has been discussed in detail by Evans and Goodman (1941a). The determination of thorium from the thoron content has likewise been mentioned insofar as it provides additional evidence for the validity of the thorium values obtained from alpha-count measurements. Instead of repeating this material, the present discussion supplements these previous remarks with details that particularly pertain to the age determinations reported in a later section and in the paper by Hurley and Goodman (1941).

We have seen in the preceding section that a negligible experimental error in the determination of the rate of helium production is introduced by the lack of knowledge of F for alpha counts on rock deposits of about 5 micra in thickness. Since the area is about 40 cm², such sources only represent a mass of 60 mg of rock. Except for quite homogeneous rocks, a single measurement on this small quantity of sample cannot be expected to give a very representative average value for the radioactive content. Consequently, it is customary to grind between 1 and 2 grams of sample from which several deposits are made for alpha counts. The average value is then used for the age calculation. This procedure gives quite satisfactory results unless the activity index of the sample is unusually low—*i. e.*, less than 0.4. Below this activity, the radon-thoron method is definitely superior to the alpha-count method. For this reason both methods are in active operation at the Massachusetts Institute of Technology laboratories and supplement each other in covering the entire range of activities found in terrestrial materials.

The thoron method being used at present is similar to that described by Urry (1936, p. 35) but embodies a number of changes in technique that appear to improve the reproducibility of the results. Since Urry (1933, p. 335) employed a sodium carbonate fusion for releasing helium from the rock sample, he used the same flux for radium and thorium as well. For inhomogeneous rocks, this procedure offers a distinct advantage in using the same portion of sample for all three measurements. A pyrex, bell-jar furnace suitable for carbonate-fusion helium measurements is shown in Figure 3. Instead of the thimble-shaped, nickel crucible used by Urry, a hemispherical nickel crucible, 5.0 cm in diameter, is used, because it allows more rapid solution of the melt following fluxing. The use of sintered-glass filter plates with suction greatly accelerates the otherwise very laborious chemical manipulations and

reduces the uncertainty concerning the time of separation of the thorium X from its parent, radiothorium. It has also been found that nitric acid solutions of thorium X have a higher and more reproducible emanating power than corresponding hydrochloric acid solutions. For this reason,



THORIUM APPARATUS

FIGURE 7.—*Diagram of thorium apparatus*

the procedure of dissolving the final residue, after removal of silica, in 50 cc of dilute nitric acid has been followed.

This solution is then placed in a 75 cc flask and the thoron measurement begun using the apparatus represented in Figure 7. Radioactively inert nitrogen [14] is bubbled at a constant rate, as indicated by the tapered glass flow meter, through the solution and continuously sweeps the thoron into the ionization chamber. The gases are then exhausted through an upper outlet tube, but a definite fraction of the thoron atoms disintegrate in the chamber (Evans, 1935, p. 109-112). The ionization created by each alpha ray from thoron and its decay products is collected by the electrostatic field between the center wire and the shell of the chamber. The resulting pulses are amplified by the electrometer tube (FP-54) and fed to a sensitive galvanometer. The position of the galvanometer is followed photographically. Since the 6″ by 37″ bromide paper used for recording moves at a known rate, the alpha particle emission rate in alphas per

[14] Ordinary tanked nitrogen, stores for 30 days or longer to allow any radon present to completely decay, has been found quite satisfactory.

hour is obtained directly from a visual count of the developed record. When the emission rate is plotted against the time, a histogram similar to Figure 8 is obtained. It is seen that the rate increases abruptly above the normal background activity upon introduction, at the 20 hour mark,

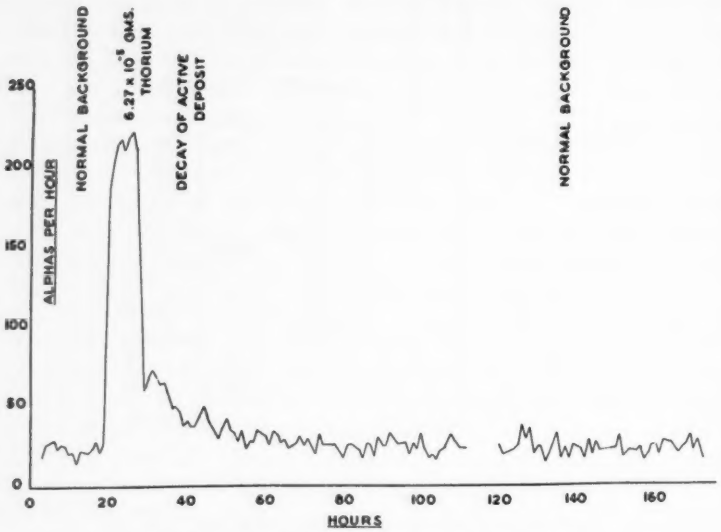


FIGURE 8.—*Thorium calibration and background runs*

of a solution containing thorium X, in this case 15 cc of our temporary thorium standard. (*See* Evans and Goodman, 1941a, p. 469-470). The average activity for the 8 hours during which streaming continued is found to be 190 alphas per hour above a background of 22 alphas per hour. Other calibrations over the range of activities observed for most rocks give the curve shown in Figure 9.

It should be noted from Figure 8 that the background following the rock run does not fall immediately to the normal value. Instead there is clearly a gradual decrease over a period of 50 to 60 hours after removal of the thorium X solution. This activity is attributable to the active deposit of thorium B[15] collected on the central electrode of the ionization chamber. This element decays with a half-period of 10.6 hours, which is much longer than that of the other daughter products of thorium X. Hence, the rate of decay of the residual deposit activity is dictated by

[15] The genetic relation between thorium B and the other members of the thorium series can be recalled by reference to Figure 1.

thorium B. In practice it is quite inefficient to wait 50 to 60 hours between runs, yet accurate knowledge of the background is essential for reliable measurements. Accordingly, the procedure is to make extended

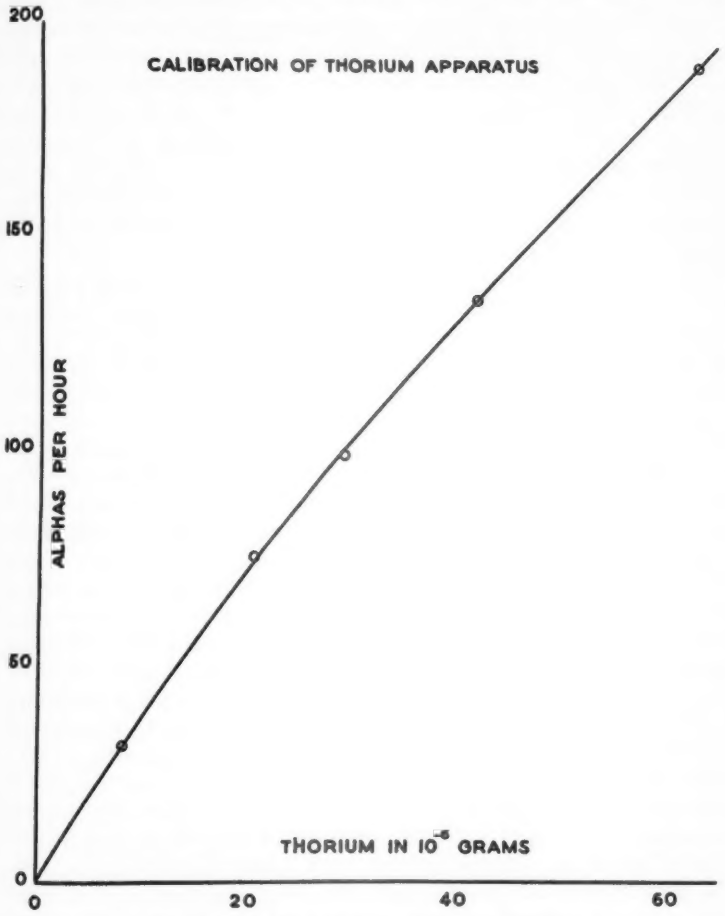


FIGURE 9.—*Calibration curve of thorium apparatus*

determinations of the normal background every 5 or 6 runs and observe the decaying background for 16 hours after each run. Knowing the decay rate, the true average background for each rock measurement can be computed from the combined observations. This schedule makes possible

one thorium measurement or calibration run every 24 hours during the week and an extended normal background measurement over the week-end. The most important desideratum is to make the calibration, rock measurement, and background runs all under as close to identical conditions as possible. Toward this aim the above schedule is followed for both calibration and rock measurements, and the same procedure followed for backgrounds with distilled water substituted for the thorium X solution. Under ordinary conditions, the moisture picked up from the solution in the flask would condense to some extent in the ionization chamber and cause electrical leakage across the amber insulation of the center electrode. A few turns of resistance wire placed around the chamber and dissipating 5 to 10 watts raises the temperature in the chamber sufficiently to prevent this condensation.

After completing the thorium measurement, the same solution can be used for a radium determination using the procedure for liquid solutions described by Evans (1935, p. 103). In brief, the procedure is: (1) transfer the solution to another flask, (2) seal this flask to one of the reflux condensers shown in Figure 3, (3) de-emanate the solution by heating, while bubbling through radioactively inert nitrogen, (4) shut off for a known period of time (usually several days) during which a known fraction of the total radon is produced by the radium in the solution, and (5) transfer this radon to the ionization chamber and measure the rate of alpha emission by means of the apparatus shown in Figure 3. Calibration with standard radium solutions (*see* Evans and Goodman, 1941a, Figure 1) following the same procedure, gives the conversion to equivalent radium content. The main difficulty with the procedure is the uncertainty due to radium contamination from the chemicals and glassware used in the fluxing, purification, and dilution of the rock sample. Even with very elaborate precautions, it has been found very difficult to avoid picking up between 0.2×10^{-12} and 0.3×10^{-12} grams of radium in the over-all procedure, as indicated by blank measurements. This correction, which is not always reproducible, becomes very significant for rock samples of low radium content and may even exceed the radium content of 5 grams of sample. Accordingly, it is preferable to limit this method to rocks with higher than average radium contents and make direct-fusion measurements on separate portions of sample for the less active rocks. The direct-fusion furnace technique avoids all these sources of contamination in radium measurements.

PRIMARY HELIUM

Rayleigh (1938, p. 410) and Shepherd (1938, p. 311) have established the presence of the noble gases, notably argon, in rocks. Although there is a possibility that some argon may be produced from the disintegration

of potassium, as shown in a later section, the major proportion of these inert gases is very likely of primary origin. Since magmas are known to contain uranium and thorium, they must also contain some helium due to radioactive disintegration. If other inert gases in rocks are derived from the magma, it is only reasonable to suppose that at least a part of the helium content of some igneous rocks must be of a similar origin. If primary helium constitutes a significant fraction of the observed helium content of a rock, the third requirement is not fulfilled and the age calculated from Equation (1) will be in error on the high side.

At first sight, it would seem that helium measurements on *recent* effusive rocks would provide a direct means of ascertaining the amount of primary helium. However, the texture of these rocks vitiates their usefulness. As the result of their mode of origin, effusives are generally either glassy or porous. Since it is well established (Paneth and Peters, 1928, p. 253; Urry, 1932, p. 3887; 1933, p. 3242; Rayleigh, 1936, p. 350) that many glasses are selectively permeable to helium, and since the atmosphere contains helium [16], glassy, effusive rocks are likely to contain sufficient atmospheric helium to mask any contribution from their magmatic sources. The helium content of a number of recent Vesuvian lavas, as observed by Strutt (1907, p. 436), is probably of atmospheric derivation. Other more porous effusives have been found by the authors to contain neon in the proportion of about three parts to one part of helium by volume, which suggests that air is either absorbed or entrapped in the rock.

The information obtained from studies of recent trap rocks is of considerably more value in studying the question of primary helium. The absence of air from the environment of the rock at the time of its intrusion eliminates the possibility of absorption of atmospheric helium. However, helium measurements alone are not sufficient to evaluate the presence of primary helium, for it is not possible to obtain specimens of these intrusive rocks that are geologically very recent. In addition, the helium content of rocks, even of the same age, varies over a wide range owing to the considerable variation in the radioactive content, and hence of the rate of production of helium, as shown by Evans and Goodman (1941a). For this reason, it is necessary to measure the radioactivity of the rock as well as its helium content. These data combine, according to Equation (1) to give the age of the rock. Thus we see that helium ages themselves provide the most direct means of studying the question of primary helium. But the term, age, can justifiably be applied only when the four fundamental requirements are rigorously fulfilled. Hence,

[16] In a recent compilation, Paneth (1937, p. 436) gives the following percentages for the rare gas constituents of air: Ne: 1.8×10^{-3}, He: 5.3×10^{-4}, Kr: 1×10^{-4}, H_2: 5×10^{-5} and Xe: 8×10^{-6}.

TABLE 1.—*Age measurements of Tertiary dike rocks*

Sample No.	Rock specimen	Helium-content in 10^{-3} cc/g	Activity index		Age ratio in 10^6 years
			Alpha method	Rn-Tn-Method	
4B	Giant's Causeway[1]	*0.31±0.03	**0.44±0.03	.....	22. ±4
5I	Cleveland Dike[1]	0.39±0.03	0.7 ±0.1	.....	17. ±3
37B	Tower Falls basalt[1]	0.09±0.03	0.50±0.03	.....	5.5±1.5
....	Douglas Creek basalt[2]	*0.22±0.03	.....	*0.42±0.04	<16. ±1.0
....	Columbia River basalt[2]	0.27±0.03	.....	0.46±0.04	<18. ±1.4
....	Mt. Pawagarh plug[3]	*0.11	.....	*0.68	5.
....	Mt. Pawagarh basalt[3]	0.12	.....	0.10	37

[1] Geologic notes for these specimens are given in a later section of this paper.
[2] These age determinations by Urry (1936, p. 47) represent maximal values due to errors in the radioactivity measurements.
[3] Early determinations by Dubey, reported in National Research Council, 1931, p. 413.
* Mean of two determinations.
** Mean of four determinations.

in this circuitous method of evaluation, in which helium ages are used to test one of the requirements on which helium ages are based, one must assiduously avoid the pitfall of reasoning in a circle.

First, we must assume that we have some qualitative notion of geological time. Suppose we regard 50 million years as a relatively short span in geological time. (There is considerable independent evidence supporting this generalization.) Secondly, we must assume that by selection of samples we are able to evaluate one uncertainty at a time. This is necessary, since if primary helium is present and helium leakage has occurred subsequent to solidification of the rock, the two effects are mutually compensating, and it is not possible to evaluate either on a given specimen. Suppose for the moment we assume that fresh, fine-grained, basic igneous rocks do not allow helium to diffuse either into or out of themselves. Then age ratios on recently formed intrusives of this type should give some indication of the presence or absence of primary helium.

Measurements have been made on a number of such Tertiary rocks, and the results are presented in Table 1. It is seen that the observed helium contents are quite small. Even if all this helium were of primary origin, it would be of negligible amount compared to the radiogenic helium that would be produced in these rocks within a few hundred million years by the radioactive contents observed. Hence, corresponding amounts of primary helium in much older rocks would not significantly affect the measured helium age, except in cases of exceptionally low radioactivity. Since we have every reason to believe that the specimens in Table 1 are typical, they furnish fairly definite evidence that for trap rocks at least the amount of primary helium, if any, is usually negligible in helium age measurements. It remains for future research to evaluate this question for the more plutonic rocks.[17]

MIGRATION OF HELIUM AND OF RADIOACTIVE ELEMENTS

General considerations.—We now consider the fourth and final requirement of the helium method—namely: has there been any addition or subtraction of helium or of the radioactive elements during the history of the material whose age we are attempting to measure? Since the preceding three requirements can be fulfilled simultaneously for certain selected rock samples, the fate of the helium method appears to hinge on the fulfillment of this fourth requirement.

Following the establishment of the reliability of the analytical tech-

[17] There is some evidence that primary helium occurs in certain of the Sudbury rocks, which may explain to some extent the anomalous helium age ratios observed (Hurley and Goodman, 1941, p. 546). However, the data are too limited to draw any definite conclusions, and in addition these rocks are not typical of plutonics in general.

niques, it became increasingly apparent that only after a thorough study of the fourth requirement had been made would a true evaluation of the helium method be possible. Accordingly, in the Fall of 1939, a systematic investigation of the problem of helium retention in rocks and minerals was initiated in collaboration with Dr. P. M. Hurley of the Massachusetts Institute of Technology Geology Department. Although the problem of the migration of the radioactive elements in rocks is far from settled, the question of possible helium migration seemed to be the more uncertain of the two aspects of the fourth requirement. For this reason, study of the latter has been undertaken first. It is in the sampling of the proper materials by the geologist for analysis by the physicist that the co-operative aspects of such research becomes of paramount importance. Our present knowledge of the fourth requirement is the sum of the following inventory of information and the results of the recent work reported in the paper by Hurley and Goodman (1941).

Helium migration.—A number of laboratory studies have been made of the diffusion of helium in rocks and minerals. Urry (1933, p. 3242) and Rayleigh (1936, p. 350) each measured the permeability to helium of one rock specimen and a number of minerals. Because of the minute quantities of transmitted helium, relatively large pressures were required. The general nature of the results indicates a very low permeability. However, extrapolation to geological conditions involves so many uncertainties that these observations only afford qualitative evidence for the retention of helium over geologic expanses of time.

The fractional loss of helium at elevated temperatures has been studied for a very limited number of meteorites and rocks and for a more extensive, but for our purpose less important, number of radioactive minerals. Paneth and Urry (1930, p. 140) have demonstrated that the loss of helium, from three iron meteorites maintained at 1000°C for several hours, was equivalent to the helium contained in only a thin surface layer. Such meteorites show the greatest retention of helium of all mineral matter studied, with the possible exception of certain of the native metals.

The escape of helium as a function of grain size and time has been studied for a few rock samples. One 8-gram sample of each of two fine-grained specimens, a basalt and a felsite, were found by Urry (1933, p. 335) to lose less than 0.05 and 0.03 per cent respectively of their helium per 100 years, based on measurements made after being sealed in air-free oxygen at atmospheric pressure for 3 months. Urry also found no difference in the helium content of a Keweenawan basalt, whether taken in one piece, several small pieces, or finely powdered, after standing for 24

hours between grinding and fluxing. In contrast to these results, Holmes and Paneth (1936, p. 385) observed a 60 per cent loss of helium from finely powdered kimberlite kept in a glass tube with a cork stopper at atmospheric pressure for 4 years. Too much emphasis should not be placed on these measurements since the helium leakage from powders is only of real significance in evaluating the analytical techniques. It is to be expected that considerable helium might escape from a rock powder prepared from a rock which in its native state would retain its helium completely. Truly significant measurements of this type should be made on specimens of at least hand size.

The most elaborate measurements thus far made on the helium retention of rocks and rock-forming minerals are those reported by Holmes and Paneth (1936, p. 385). Although these observations are quite complete from an analytical standpoint, it is to be regretted that this excellent work had to be done on specimens which are so atypical of average igneous materials. The excessive hydrothermal alteration of the various rocks composing the Kimberley diamond pipes indicates clearly that such rocks cannot be expected to yield anything approaching reliable helium ages. The relatively high helium contents of certain of the basic appinites and eclogites are significant only in that they indicate retention of a large fraction of their helium under the most adverse geological conditions. The phlogopite glimmerite and basic granulite are so highly altered that helium loss from these specimens can only represent extreme conditions compared with those to be expected for typical igneous rocks.

For the sake of completeness, some mention should be made of the loss of helium from radioactive minerals, which in general contain about a millionfold more helium than ordinary igneous rocks (Fig. 2). In his early work on the age of such minerals, Strutt showed that there is an appreciable loss of helium even at room temperature. Kano and Yamaguchi (1928, p. 244) have studied the yield of helium from samarskite and ishikawaite, with and without the addition of flux, when held under vacuum for 2 hours at temperatures from 200 to 1000°C. These investigations also indicate that more than 90 per cent of the helium is lost from ishikawaite during the first 30 minutes when held at 700°C for 8 hours. Time intervals shorter than 30 minutes were not used. Other researches (Chlopin, Herling, and Joffe, 1934, p. 28) have shown that the release of helium from radioactive minerals is greatly accelerated by the presence of hydrogen in the surrounding gases. Since hydrogen very seldom occurs in nature, its influence would be rare, although this fact may have an important bearing on the genesis of helium-bearing natural gases.

A piece of work [18] particularly interesting since it was made only one year after the discovery of helium on the earth was carried out by Ramsay and Travers (1897, p. 325). They found that fergusonite, when heated to a temperature not exceeding 500°C, becomes suddenly incandescent and evolves much of its helium, while its density decreases. The investigators thought at that time that the effect was due to "breakdown of a helium compound" and they called fergusonite an "endothermic mineral." X-ray study in recent years has shown that fergusonite normally has not a crystalline atomic structure but on heating to 400°C it becomes crystalline, tetragonal. This suggests the possibility that a temperature condition, which causes a state of dynamic disorder either among part or among all of the constituents of a crystal structure, might release part of the helium held in the structure, even if the disorder is only momentary.

With modern knowledge of crystal structure, it should be possible to make a theoretical estimate of the probabilities of diffusion of helium through minerals. The most important assumption involved in such considerations is that of assigning the distribution of the radioactive elements which generate the helium. Practically no definite information is available concerning this interesting problem. In the absence of experimental evidence it is generally assumed that the uranium and thorium atoms are randomly distributed throughout the crystal structure of the rock mineral. On this basis Keevil (1940, p. 311) has recently shown theoretically that a surprisingly large retention of helium should exist for igneous rocks which have not been subjected to hydrothermic or dynamic metamorphism. Feldspar, in particular, is considered in detail, and Keevil arrives at the conclusion that this mineral, when fresh, should not lose an appreciable fraction of its helium during a length of time comparable to the age of the earth itself. Hurley and Goodman (1941) present experimental evidence which indicates quite the contrary in the case of feldspar. Accordingly it must be concluded that this theoretical treatment does not adequately include all factors involved.

As is so frequently the case, laboratory facilities cannot duplicate geological conditions, and in order to evaluate the fourth requirement reliably we must base most of our conclusions on field evidence. By the judicious selection of certain critical specimens, it is possible from age measurements to make important deductions concerning the retention of helium in rocks. If there is any appreciable loss or migration of helium either in the rock body as a whole or in the individual mineral constituents of the rock, different values for the helium age ratio should be obtained, since the helium would no longer be in immediate association

[18] The authors are indebted to Dr. P. M. Hurley for calling their attention to this early work.

with the corresponding amount of the parent radioactive elements that produced it. In the course of his extensive age measurements of the Keweenawan, Urry (Lane and Urry, 1935, p. 1101) selected a few rocks for their bearing on this question of helium retention. These measurements appeared to afford convincing evidence of the lack of migration of helium. However, as has been mentioned previously, the radium measurements obtained in these studies were too low by a factor of more than two. Because of the variations in the thorium-uranium ratio of these specimens the downward revision of age ratios results in widely discordant values, with the result that these critical measurements on helium retention must be discarded entirely.

The variation of the helium age ratio, laterally or with depth in the same igneous body, would suggest the migration of helium. Two of the early measurements by Dubey [19] on acidic felsite of the Mt. Pawagarh plug indicated the same age ratio for a specimen from the summit and one from 100 feet below. Holmes has discounted the absolute value of these age ratios because of the cryptocrystalline nature of the rocks. In recent years Keevil (1938, p. 123) found the same age ratio over a depth of 600 feet in Quincy granite, determinations of ages having been made at five points over this range. Further age measurements, on this well-known rock, from quarries separated by a tenth of a mile and one mile, also gave the same age ratio, indicating a lack of differential migration of helium. Keevil also obtained the same age 2 feet from the margin and in the center of a Keweenawan trap dike. However, these latter results are not thoroughly convincing evidence of retention of helium since the radioactivities were essentially the same, and hence no gradient of helium existed.

Some of the first age measurements made by Dubey (1930, p. 807) on the Gwalior series gave progressively lower age ratios with an increase in grain size for three rocks that were considered to represent different phases of the same basic intrusive. These discrepancies in age were attributed to the loss of helium in coarser-grained materials. It is interesting to note that in the original publication of these results the textures of the rocks were indicated merely as fine-grained or medium-grained. Subsequent recapitulation has given three gradations of grain size, increasing in the order of decreasing values of the reported age measurements. A number of recent determinations by Goodman and Keevil, included with other results in a later section, fail to confirm this variation of age ratio with texture for basic rocks.

A number of metamorphic and sedimentary rock types are known to be unsuitable for helium age determinations because of the escape of helium.

[19] Personal communication to Arthur Holmes; *see* National Research Council, 1931, p. 413.

The major portion of the rocks reported by Holmes and Paneth (1936, p. 385) fall in the class of metamorphic rocks comprising hydrothermally altered igneous rocks. The age ratios obtained indicate extreme conditions of helium loss.

TABLE 2.—*Age measurements on metamorphosed rocks*

Sample No.	Geologic age	Helium content in 10^{-3} cc/g	Activity index in[+] α/hr•mg	Age ratio in 10^6 years
38B	Pre-Cambrian.....	*2.7 ±0.2	**0.97±0.05	85±5
4M	Pre-Cambrian.....	2.2 ±0.2	*0.82±0.05	84±8
5M	Pre-Cambrian.....	2.4 ±0.2	*2.0 ±0.1	37±3
6M	Pre-Cambrian.....	*0.33±0.04	*0.18±0.01	56±9
7M	Cambrian (?).....	*1.3 ±0.1	0.88±0.04	46±5
33A	Paleozoic.....	*0.97±0.08	**0.50±0.04	62±8
31A	Paleozoic.....	3.4 ±0.3	*2.8 ±0.1	37±3
18	Pre-Cambrian*†.....	0.43	0.84	16
20	Pre-Cambrian*†.....	3.7	1.17	98

[+] Activity index measurements by alpha count except for samples 18 and 20, where radon-thoron method was used.

† Determinations by Holmes and Paneth (1936, p. 408).

* Mean of two determinations.

** Mean of four determinations.

Two of Holmes and Paneth's results on pre-Cambrian rocks together with measurements by the authors on six geologically old rocks are given in Table 2. These age ratios are much lower than those observed for unaltered rocks of comparable geologic age.[20] The most likely explanation is that metamorphic changes cause at least a partial loss of helium from rocks. While the introduction of uranium and thorium would afford an alternative explanation, this process is much less likely, as indicated in the next section.

Migration of radioactive elements.—The migration of radioactive elements commonly occurs as the result of movement of solutions (Henderson, 1939, p. 238) in which these elements are either suspended or dissolved. Some migration occurs in the gaseous phase. Diffusion in the solid state appears to be exceedingly rare. With only a few exceptions, the important uranium and thorium minerals are formed by high-temperature replacement of earlier pegmatitic minerals. The concentrated solutions of these parent elements, that effect the replacement, represent the uranium and thorium content of large volumes of magma. These

[20] There is some possibility that the period of metamorphism may be estimated by studies of this type, but the uncertainties involved in such an application of the helium method are even more numerous than those for unaltered rocks.

solutions escape from the molten rock as the result of the filter-pressing action of the crystallized fraction of the cooling rock mass.

The migration of the parent radioactive elements into rocks, as the result of replacement, or out of rocks, as the result of leaching, will respectively decrease or increase the age ratio. The only value we can observe for the denominator of the general age equation is the present-day rate of production of helium, which, as we have seen, is proportional to the radioactive content. If any significant amount of replacement occurred in a common rock, there would be a number of indications seen upon petrographic examination. Rocks of this type should not be studied by the helium method until sufficient knowledge has been obtained regarding the importance of this effect. Similarly, fresh, unweathered specimens should always be chosen, since leaching may cause erroneous results. These considerations emphasize the need for adequate petrographic study prior to helium age measurement. This policy has only been followed in the more recent age research.

A study of the distribution of the radioactive elements over a regional scale provides a check on the possible migration of these elements since solidification of the rock mass. It is difficult to conceive of a process that would result in uniform addition or subtraction of both uranium and thorium throughout a large mass of rock. The results of a study of the Triassic intrusives of New Jersey, Pennsylvania and Virginia are given by Hurley and Goodman (1941). Good agreement is observed between age ratios for selected minerals separated from specimens of the same rock mass over a lateral range of 45 miles. These results indicate that in this rock body, at least, no significant migration of the radioactive elements has taken place since its solidification.

AGE MEASUREMENTS

General statement.—We have seen that the reliability of the helium method can be evaluated to some extent on the basis of independent physical and geological knowledge, but in the final analysis this method, or any other geological tool, must be judged by its success in actual application. The road of research is seldom smooth, and this is particularly true in the development of radioactive methods of age determination. Considerable work has been done during the past 3 decades, but much of it has been discarded as newer developments disclosed earlier misconceptions and discrepancies. Our present position can best be appreciated by a review of the past progress.

Early investigations.—Following the suggestion by Rutherford in 1905 that the accumulation of helium in radioactive minerals might provide a means of measuring geological time, Strutt made an extensive survey of

the helium, uranium, and thorium content of a number of the more radioactive terrestrial materials. He obtained helium age ratios for phosphatic deposits and hematite (1908, p. 272), other iron ores (1909, p. 96), zircons (1909, p. 298), and sphenes and other minerals (1910, p. 194). From these results a broad correlation between the geological age of a mineral and the helium age ratio was established. However, Strutt (1909, p. 166) was the first to point out that, owing to leakage of helium from all these minerals, the age ratios observed could only represent minimal estimates of the true age. These helium age ratios were found to be only 30 to 70 per cent of the values based on the accumulation of lead in corresponding radioactive minerals. For this reason, it has been necessary to discard most of the excellent helium age researches of Strutt.

Waters (1909, p. 677; 1910, p. 903) made preliminary helium age studies of some common rock minerals. Unfortunately, the electrical detection methods used were too insensitive to measure other than the more radioactive mineral inclusions. Because these materials showed evidence of helium leakage, the work was discontinued.

Following the development of methods of measuring minute quantities of helium, Paneth and his coworkers determined helium age ratios for a large number of meteorites (Paneth and Urry, 1930, p. 127). The magnitude of the age ratios obtained, together with some laboratory tests on the materials, indicates that iron meteorites have a remarkably high retentivity for helium, even at elevated temperatures. Similar measurements were attempted on native metals, but their extremely low radioactivities prevented application of the method. Subsequently, Paneth has collaborated with Holmes (1936, p. 385) on an investigation of the rocks composing the Kimberley diamond pipes. These measurements are mainly of value in emphasizing the type of material *not* to be used for helium age studies.

The first real stride in the application of the helium method to rocks can be said to have begun with the work of Dubey and his collaborators. These results, summarized in Table 3, illustrate a number of interesting points. The felsite (1) shows a much lower age ratio than the Cleveland Dike (2), although the two rocks are of approximately the same geologic age. The age ratios of (2 - 5) are in the proper sequence, but the actual values observed are somewhat lower than the corresponding lead ages. Specimens (5 - 7) were selected to test the effect of texture on the retention of helium as well as to estimate the geologic age of the Gwalior series of India. In the original publication Dubey (1930, p. 807) listed (5) as a fine-grained basalt and both (6) and (7) as medium-grained dolerite from a Morar-group sill. Holmes, (National Research Council, 1931, Table LXXVI, p. 415), presumably on the basis of a more exact

description, subsequently indicates (6) as medium-grained and (7) as "of coarser grain than 6." With this revision in the texture, it is seen that the age ratios for (6 - 8) decrease with increasing grain size. In the light of these observations, it seemed desirable at that time to concentrate on close-grained, crystalline basaltic or other basic rocks.[21]

TABLE 3.—*Age measurements of igneous rocks*

Rock specimen	Geologic age	Helium content in 10^{-4} cc/g	Activity index Rn-Th method	Age ratio in 10^6 years
1. Acid felsite	Miocene	*0.11	*0.68	5
2. Cleveland Dyke	Oligocene or Miocene	0.11	0.12	28
3. Deccan basalt	Early Eocene	0.12	0.10	37
4. Whin Sill	Late Carboniferous	0.36	0.057	196
5. Gwalior basalt	Late pre-Cambrian	0.55	0.034	500
6. Morar dolerite	Late pre-Cambrian	0.42	0.033	389
7. Morar dolerite	Late pre-Cambrian	0.31	0.047	203

1-7 Geologic notes, references and further discussion, National Research Council, 1931, p. 411-415.
* Mean of two determinations.

Following this work, researches were initiated in 1931 by Dr. W. D. Urry in collaboration with Professor A. C. Lane on an extensive suite of Keweenawan rocks and minerals,

"with a view to co-ordinating age results with certain geological and mineralogical aspects pertaining to the specimens, in addition to the important question of the age and duration of the Keweenawan" (Lane and Urry, 1935, p. 1119).

The magnitude and scope of this work were indeed admirable. As subsequent events have proved, however, it was premature. Age ratios were obtained for more than 30 rock specimens and minerals. With only a few exceptions, excellent agreement with geologic conditions was observed. The magnitude of the age ratios also appeared to be in accord with equivalent lead ages. On the strength of this encouragement age ratios were determined on 39 selected rock specimens ranging in geologic age from Cambrian to Pliocene. Urry (1936, p. 1226) reports that of these results

"only five are incompatible with the correct geologic sequence as determined from field relations. All five specimens should be rejected as unsuitable on strict adherence to the standard conditions set for selecting material."

The helium time scale represented by the summation of these measurements was also found to agree quite well with the somewhat less extensive

[21] Holmes (National Research Council, 1931, p. 414) states as follows: "A systematic programme of research has already been started by Dr. Dubey in India and by the writer in collaboration with Prof. Dr. F. Paneth, and Dr. R. W. Lawson who have arranged to make helium determinations and to investigate quantitatively the escape of helium under various conditions." Unfortunately these investigators have found it possible to complete only partially this admirable program.

TABLE 4.—*Helium age ratios of basic igneous rocks*

Sample No.	Rock specimen	Location	Geologic age	Age ratio in 10^6 years
1B	Porphyritic basalt	Yellowstone Park	Tertiary	5
37B	Tower Falls basalt	Yellowstone Park	Recent	5.5 ± 1.5
3B	Crescent Hill basalt	Yellowstone Park	Tertiary	9 ± 1
I	Overhanging Cliff	Yellowstone Park	Miocene	11 ± 4
5I	Cleveland Dike	Durham, England	Oligocene or Miocene	17 ± 3
4B	Giant's Causeway	England	Lower Tertiary	22 ± 4
2	Deccan basalt	Mt. Pawagarh, India	Early Eocene	37
7B	Geode Creek basalt	Yellowstone Park	Tertiary	36 ± 4
1	Kimberlite	South Africa	Late Cretaceous	58
6B	Oldwick basalt	Oldwick, N. J.	Triassic	57 ± 3
J	Dike 3,200′ level	Eustis Mine, Quebec	Paleozoic (?)	54 ± 8
113	Earlier dike	Eustis Mine, Quebec	Paleozoic	81 ± 10
27B	Monchiquite dike	Kilchattan, Scotland	Post-Devonian	83 ± 8
26B	Monchiquite dike	Riasg Buidhe, Scotland	Post-Devonian	105 ± 6
3	Whin Sill	Westmoreland, England	Late Carboniferous	196
32B	Dalhousie andesite	Nova Scotia	Devonian	160 ± 25
43B	Gogebic dike	Michigan	Late pre-Cambrian	190 ± 25
$24B_1$	Long Lake dike	Montana	Late pre-Cambrian	290 ± 35
14B	Noranda diabase	Quebec	Post-Timiskaming	250 ± 20
4	Gwalior basalt	India	Late pre-Cambrian	500
R	Basalt	Ropruchei, Karelia	Pre-Cambrian	600 ± 35
25B	Beartooth trap	Montana	Pre-Cambrian	1050 ±100
22B	Stillwater norite	Quad Creek, Montana	Early pre-Cambrian	1830 ±250

Analysts: Except for the following samples, the measurements were made by the authors: Dubey, *et al.* (Nat. Res. Council, 1931, p. 413-415) 2, 3 and 4; Keevil (1938, p. 147): I, J, 113, R; Goodman and Keevil (Evans, *et al.*, 1939, p. 942): 6B, 14B, 22B, 24B, 26B, 27B and 43B; Holmes and Paneth (1936, p. 400): 1.

Notes: I. Another specimen (10B) of this rock mass analyzed by the authors showed evidence of a high proportion of neon present. The age ratio, including neon and helium, was 375 ± 35 m.y. This neon may originate in air absorbed by the rock at the time of solidification.

5I. An earlier determination by Dubey and Holmes (1929, p. 794) on a separate sample of this rock gave an age ratio of 28 m.y.

3. Recent measurements by the authors on a separate sample (7I) of this rock gave a substantially higher age ratio than that observed by Dubey and Holmes. However, there is evidence of extreme inhomogeneity in the distribution of radioactivity in this rock, and the sample at hand was too small to allow check measurements to be made.

5I and 3. These two rocks are intermediate in composition between acidic and basic but have been included in the present table because the age ratios observed agree more closely with basic rocks than with acidics.

lead time scale. This mutual compatibility of the two radioactive methods of age determination appeared as the final conclusive proof of the reliability of both methods when properly applied. While there were a number of gaps left in the structure and much work remained to be done in the pre-Cambrian, in general, the geologic time scale seemed a definitely established framework on which future research would build.

Concurrent with these developments (1929-1936), Evans was engaged in the development and application of sensitive detection instruments for the measurement of terrestrial radioactivities. (*See* Evans, 1935, p. 99-112, for key references.) The logical extension of this work was in the direction of age measurements. The physical methods used offered a number of analytical advantages that should make analyses more accurate and more routine and, hence, more generally available for geologic problems.

Intercalibration measurements.—In February 1936 collaboration between the authors began on helium age studies. The first step taken was to test the reliability of the alpha-counting method of determining radioactivity and the direct-fusion method of measuring helium, using portions of hand samples of rock previously analyzed by Urry. These preliminary determinations, on samples 19B, 34B, and 3I, gave age ratios substantially lower than those obtained by Urry. The differences appeared to be primarily due to a lack of agreement on the radium contents. However, it also appeared possible that the discrepancies in radium results might be due to inhomogeneities in the samples. Accordingly, a set of Keweenawan rocks, 39B to 43B inclusive, were measured by the alpha-helium method. These age ratios were in even more drastic disagreement with Lane and Urry's (1935, p. 1108) extensive list of ages for the Keweenawan. Thus it seemed advisable to make a number of direct comparisons between the alpha-helium and the radon-thoron helium methods. Portions of the same granulated rock samples and of the standard solutions used for calibration purposes were measured independently in the two laboratories.[22] The results of this work already have been summarized in the preceding general discussion. For the purposes of the present review, we need only repeat that, because of apparently unsystematic errors in the radioactivity measurements, all of Urry's helium age ratios had to be discarded.

Summary of age data.—The helium age data remaining after this drastic, but necessary, re-evaluation are summarized in Tables 4 and 5.

[22] A detailed description of this intercalibration and comparison has been given by the participants, Evans, Goodman, Keevil, Lane, and Urry (1939, p. 931-946).

TABLE 5.—*Helium age ratios of acidic igneous rocks*

Sample No.	Rock specimen	Location	Geologic age	Age ratio in 10^6 years
5	Mt. Pawagarh felsite	Kathiawar, India	Lower Eocene	5
28A	Obsidian Cliff	Yellowstone Park	Tertiary	8± 1
23A	Rhyolite	Yellowstone Park	Tertiary	22± 3
15A	Fitchburg granite	Fitchburg, Mass.	Carboniferous	45± 5
Q	Quincy granite (series)	Quincy, Mass.	Mississippian	66± 2
$4A_1$	Chelmsford granite	Chelmsford, Mass.	Mississippian	72± 6
11A	Granite	Franklin, Maine	Middle Paleozoic	60± 6
7A	Rockport granite (fine)	Cape Ann, Mass.	Middle Paleozoic	88± 5
8A	Rockport granite	Cape Ann, Mass.	Middle Paleozoic	100± 8
NB5	Hampstead granite	Hampstead, N. B.	Devonian	109± 5
Ga	Granite	Gananoque, Ont.	Late pre-Cambrian	150± 20
17A	Kingston granite	Kingston, Ont.	Late pre-Cambrian	280± 30
4I	Granodiorite	E. La Motte Tp., Que.	Late pre-Cambrian	195± 15
T2A	Granite	Taschereau, Que.	Late pre-Cambrian	250± 20
6I	Granodiorite	Varsan, LaCorne Tps., Que.	Pre-Cambrian	60±100

Analysts: Dubey (Nat. Res. Council, 1931, p. 413): 5; Keevil (1938, p. 146): Q, NB5, Ga and T2A; Goodman and Keevil (Evans, *et al.*, 1939, p. 938-942): remaining samples.

Notes: Q—Age measurements in good agreement with the above value have also been made by the authors on a separate sample (12A) of this well-known rock.

4I and 6I—Intermediate in composition between acidic and basic.

The values given include the earlier results of Dubey and Holmes from Table 3 and one measurement by Holmes and Paneth. The remaining age ratios include those obtained in connection with the interchecking work mentioned above, other concurrent measurements by Keevil (working in Urry's laboratory), and subsequent determinations by the authors. The helium age ratios fall into two groups, one for basic rocks (Table 4) and one for acidic rocks (Table 5). These have been placed in their appropriate positions on the left hand side of the time scale shown in Figure 10. The field evidence, on which the geological age is based, is in some cases only very approximate, and in these instances the position assigned is the midpoint of the range within which the age can be defined. The lead ages shown on the right side are also divided into two groups, one for lead ratios and one for lead isotope ratios. These two methods yield results that are in general agreement, although there are notable exceptions, for example the Permian pitchblende from Bohemia and the Cambrian Swedish Kolm. These lead age methods and the results obtained by their application are discussed in some detail in the next section. For our present purpose, the important generalizations to be drawn from Figure 10 are: (1) With only a few exceptions the helium age ratios lie in the proper geologic sequence when the results for acidic rocks and for basic rocks are considered separately, (2) the age ratios for acidic rocks are consistently lower than the age ratios for corresponding basic rocks, and (3) the helium age ratios, even for the basic rocks, are substantially lower than corresponding lead ages on radioactive minerals. These three general inconsistencies in helium age ratios indicated quite clearly that there was some fundamental failure in the helium method. A suggestion of the possible difficulty had come from Keevil's (1938, p. 406-416) investigation of the Quincy granite. Keevil observed no lateral or vertical variation of age ratio in this rock body, but he did find marked differences in the age ratios for the three main mineral components of this rock. A theoretical treatment of the probability of helium leakage from minerals (Keevil, 1940, p. 311) with particular consideration of feldspar, which was the Quincy rock mineral that gave the lowest age ratio, led Keevil [23] to conclude that the helium retentivity of practically all igneous rock minerals is sufficient to ensure the retention of essentially all the helium produced in them during geologic time. On the strength of this conviction, Keevil (1938, p. 406) attributed the observed differences in age ratios to leakage of helium resulting from alteration of the feldspar (chiefly microperthite). He states (p. 412): "if the feldspar crystals were perfect, no loss of helium could occur, and even with a normal amount of

[23] This investigation was made in 1937 although its publication was delayed until 1940.

HELIUM METHOD Igneous rock age ratios in millions of years					LEAD METHODS Radioactive mineral age ratios in millions of years		
Acidic rocks		Basic rocks			Lead ratios		Isotope ratios
Mt. Pawagarh felsite	5	Porphyritic basalt	5				
Obsidian Cliff	8	Tower Falls basalt	5				
Yellowstone rhyolite	22	Crescent Hill basalt	9				
		Overhanging Cliff	11	TERTIARY			
		Cleveland Dike	17				
		Giant's Causeway	22		34	Uraninite, Mexico	
		Deccan basalt	37			Brannerite, Idaho	
		Geode Creek basalt	36				
		Kimberlite	58	CRETACEOUS	70	Pitchblende, Colorado	
				JURASSIC	123	Ishikawaite, Japan	
		Oldwick basalt	57	TRIASSIC			
				PERMIAN	220	Thorite, Norway	
Fitchburg granite	45	Whin Sill	196			Jachymov, Bohemia	140
Quincy granite	66			CARBONIFEROUS	232	Uraninite, N. Carolina	
Chelmsford granite	72	Monchiquite dike	83				
		Monchiquite dike	105				
Franklin granite	60						
Rockport granite (fine)	88	Eustis Dike	54	DEVONIAN	269	Pitchblende, Silesia	
Rockport granite	100	Eustis Earlier dike	81		278	Various Minerals, Conn.	
Hampstead granite	109	Dalhousie andesite	160				
				SILURIAN			
					349	Uraninite, Mass.	
				ORDOVICIAN		Cyrtolite, New York	300
					366		375
					371	Uraninite, Conn.	
Gananoque granite	150	Gogebic dike	190				
Kingston granite	280	Long Lake dike	290	CAMBRIAN	400	Kolm, Sweden	770
La Motte granodiorite	195	Noranda diabase	250		580	to 995 Pitchblende, Katanga	610 to 665
					531	Ceylon Thorianite	485
Taschereau granite	250	Gwalior basalt	500				
		Ropruchel basalt	600		765	Besner uraninite	825
					803	Morogoro uraninite	595
Varsan granodiorite	860				882	Pied des Monts cleveite	905
		Beartooth trap	1050		900	Broggerite, Norway	
					1085	Norwegian cleveite	1090
					1077	Wilberforce uraninite	1035
		Stillwater norite	1830				
					1251	Great Bear Lake pitchblende	1420
					1500	Uraninite, S. Dakota	
					1570	Huron Claim uraninite	2200
					1850	Uraninite, Russia	

FIGURE 10.—*Radioactive time scales*

crystal imperfection, the permeability would be low." However, Keevil did suggest that if further work on fresh minerals substantiated these observations (p. 416), "absolute helium age determinations will be obtained only from certain minerals and not from the rock as a whole."

Recent developments.—In view of these numerous uncertainties in the applicability of the helium method, it appeared essential that a really fundamental investigation be made of the basic requirements underlying this method. The preceding review of our present knowledge concerning the first three requirements constitutes a part of this work. The progress made to date in collaboration with Dr. P. M. Hurley on the evaluation of the fourth requirement is summarized in the paper by Hurley and Goodman. Definite differences in helium retentivity have been found for various common rock minerals. These differences offer a very logical explanation for the variations in age ratios observed for basic and acidic rocks, owing to the higher feldspar content of the latter. It can also be stated with considerable surety that all helium age ratios on rocks containing feldspar as an essential mineral are too low owing to the leakage of helium from this mineral. Since all the acidic rocks in Table 5 and most of the basic rocks in Table 4 contain appreciable proportions of feldspar, the large majority of these age ratios must be considered only as minimal values. Hence, these results are of little use in the establishment of a helium time scale or for correlation purposes. However, there is a possibility that the helium retentivity of even the more permeable rock minerals can be established as a fairly specific quantity. If this can be done, a knowledge of the mineral composition of some of the rocks previously studied may enable estimates to be made of the true helium age. Until this investigation has been made, it appears necessary to include in the helium time scale only those measurements made on selected rock minerals. This restriction limits the acceptable age ratios to those reported by Hurley and Goodman. Fortunately, the suitable minerals appear to be rather widely distributed in igneous rocks. Magnetite, in particular, offers considerable promise. Age-ratio measurements made on magnetites from widely varying horizons appear to be in a sequence consistent with geologic knowledge and in quite good agreement with corresponding lead ages. By the continuance of this work, it is anticipated that a new helium time scale can be established on a sound scientific basis. Although complicated by the necessity for greater selection in the choice of samples, the new helium method should provide a valuable aid to geologists in the solution of the numerous important problems awaiting a reliable dating method for rocks.

FIGURE 10.—*Radioactive time scales*

LEAD METHOD

APPLICABILITY

By reference to Figure 2, the fundamental difference between the helium method and the lead method of measuring geologic time has been discussed. It is evident that with present analytical techniques the lead method must be restricted, almost exclusively, to the study of radioactive minerals. In the present section an attempt is made to evaluate the extent to which the second, third, and fourth basic requirements for reliability are fulfilled by radioactive minerals. These considerations seem particularly appropriate at a time when lead ages are being used as indubitable points of reference on the geologic time scale.

ANALYTICAL TECHNIQUES AND ACCURACY

The quantitative determination of Pb, U, and Th in radioactive minerals can usually be made with an uncertainty of only a few per cent. The precision of chemical separations and gravimetric measurements, of course, depends on the concentrations of these elements in the mineral. There is also some dependence on the chemical nature of the mineral; certain of the more siliceous minerals are particularly difficult.

The presence of lead as an impurity in the reagents may be a significant source of error unless due precautions are taken, as Wells (*see* Lane, 1936, p. 66) has point out. Some of the earlier work on Pb/U and Pb/Th ratios undoubtedly suffers from this defect. However, a more important cause of inaccuracy in these measurements was the failure to determine the amount of ordinary lead which, as indicated in Figure 2, often occurs in appreciable quantities in radioactive minerals. It is necessary to be able to identify the two types of lead, ordinary, Pb, and radiogenic, Pb*, in order that the numerator of Equation (1) accurately represent only lead generated by radioactive disintegration.

Three methods have been used for distinguishing between ordinary and radiogenic lead. These three methods—(1) atomic weight determination [24], (2) determination of the abundance of the lead isotopes mass spectrographically, and (3) a similar determination from the hyperfine structure of the lead spectrum—all depend upon the difference in atomic weight of the individual lead isotopes, Pb^{204}, Pb^{206}, Pb^{207} and Pb^{208}. Because of its convenience, accuracy, and need for only small quantities of sample, together with the fact that in some cases it gives additional information not otherwise obtainable, the mass spectographic method has largely superseded the other methods. Aston (1927, p. 224; 1929, p. 313)

[24] Since the atomic weight of ordinary lead, 207.21 (chemical scale), is substantially different from that of uranium lead, approximately 206.03, and thorium lead, approximately 207.98, the proportions of the different leads in the mineral lead can often be estimated from an atomic weight determination.

made the first isotopic abundance measurements of ordinary and radiogenic leads. Nier (1938, p. 1571; 1939, p. 153) has recently greatly improved the accuracy of this technique and has made a systematic study of quite a number of ordinary and radiogenic leads whose atomic weights had previously been determined by chemical methods. From these results it appears that the present analytical techniques are adequately accurate for the identification of lead in age measurements of the large majority of radioactive minerals.

PRIMARY LEAD

In the preceding discussion a distinction has been made between ordinary lead, Pb, and radiogenic lead, Pb*. Until recently it was assumed that these two leads represented the only types found in terrestrial material. This assumption was based on the observation (National Research Council, 1931, p. 218; Baxter, *et al.*, 1935, p. 467; 1937, p. 702) that the atomic weight of ordinary lead from a wide variety of sources was apparently constant. Nier (1938, p. 1574) has recently observed significant variations in the relative abundance of the isotopes of a number of ordinary leads. These variations, however, produce little or no effect on the chemical atomic weight of the lead, which is a very insensitive indication of isotopic composition where four isotopes are involved. These variations are much larger than can be explained by possible isotopic separation taking place in nature. Instead it seems likely that what was previously considered to be ordinary lead is itself a mixture of primal lead and varying amounts of radiogenic lead. In order to distinguish between these several forms for purposes of discussion, the following definitions are included:

Primal lead.[25]—From spectroscopic analysis of solar radiation, it appears that the atoms composing the sun are identical with those of our earth. It also appears probable that the stable atoms have remained essentially the same throughout the lifetime of our planet. Accordingly, some of the lead found in rocks and minerals today must constitute part of the original inheritance of the earth. The term "primal lead" is used to represent this form of lead. Since the isotope Pb^{204} is not produced by any known radioactive disintegration, it is considered as indicative of the proportion of primal lead. However, the isotopic constitution of primal lead is shrouded in mystery; the best approximation for the abundance of its isotopes comes from a study of very old lead minerals. Thus far the lead from the Great Bear Lake galena of pre-Cambrian age (estimated to be 1300 million years) has shown the highest proportion of Pb^{204} and is tentatively selected as representing primal lead. The lead from the Joplin, Mo., galena of Late Carboniferous age (estimated to be 230 million years) has the lowest proportion of Pb^{204} of all the ordinary leads thus far studied. Nier's (1938, p. 1574) values for the relative abundance of the isotopes of these leads are as follows:

Galena	204	206	207	208	Atomic Wt. (Chem. Scale) Physical Method	Chemical Method
Great Bear	1.48	23.59	22.64	52.29	207.218	207.206
Joplin, Mo.	1.26	27.31	20.00	51.43	207.178	207.22

[25] The general adoption of this term was suggested by Professor A. C. Lane during the Conference on Applied Nuclear Physics, held at the Massachusetts Institute of Technology in October 1940.

A comparison of the atomic weights shows the relative insensitivity of the chemical method.

Ordinary lead.—By the term "ordinary lead" is meant the lead of commerce derived from such minerals as galena, cerussite, and wulfenite. Other terms that have been used for this purpose are: "ore lead", "primary lead", and "common lead". Ordinary lead is composed predominantly of primal lead together with varying small amounts of radiogenic lead. It is the occurrence of ordinary lead in radioactive minerals that complicates the lead method of geologic age determination.

Radiogenic lead.—The lead resulting from the radioactive disintegration of uranium and thorium is designated as "radiogenic lead".

Primary lead.—The lead present in the mineral at the time of its formation. This initial time is the zero point in the age of the mineral—that is, it represents the time $t = 0$. While there is some uncertainty in the designation of this time, depending on the mode of formation of the mineral, this uncertainty is probably quite small in comparison to the geologic age of the mineral.

From the determination of the abundance of Pb^{204} in the lead separated from a radioactive mineral, it is thus possible to correct for the amount of primary lead. Nier's results show that while this correction can generally be made with satisfactory accuracy it necessarily involves some uncertainty owing to the lack of specific knowledge of the isotopic constitution of this lead. Because of the extreme difference in the geochemistry of Pb and that of U and Th, the amount of ordinary lead in radioactive minerals, except those of recent origin, is generally much less than the radiogenic lead produced in them during their lifetimes. For this reason, the uncertainty due to variations in the isotopic abundance of ordinary lead is seldom significant, although it may dictate an appreciable correction for very young minerals. These considerations emphasize the need for isotopic analysis in all lead age measurements.

ALTERATION OF RADIOACTIVE MINERALS

The fourth basic requirement as applied to the lead method involves not only the question of migration of the disintegration product, lead, but also the possible variation in the concentrations of the parent elements as the result of hydrothermal alteration. Radioactive minerals are particularly susceptible to leaching, replacement, oxidation, and reduction. Microscopic examination of radioactive minerals shows that they are invariably intensely shattered. Ellsworth (1932, p. 88) has observed that the degree of shattering in radioactive minerals increases with the uranium and thorium content, and that the rock matrix surrounding radioactive minerals is likely to be penetrated by fractures which radiate in all directions from the uranium- or thorium-bearing minerals. These effects are hardly surprising in view of the continual bombardment of the mineral and its surroundings by the energetic radioactive radiations and the atomic and molecular rearrangements which are necessitated by the finite recoil of the active atoms with each disintegration. The resulting heating effect and internal pressure of helium supplement in the development of

stresses which are sufficient to cause fractures throughout the mineral within a geologically short space of time.

These fractures afford openings through which solutions can percolate in the rocks and minerals. The resulting alteration may profoundly affect the lead ratio of the mineral. There has been considerable speculation concerning the preferential leaching of lead from radioactive minerals (Holmes, 1937, p. 172), especially thorium minerals, but at the present time our knowledge concerning this possibility is exceedingly meager. Primary uranium minerals appear to be formed under reducing conditions,[26] and it is generally assumed that all, or nearly all, the uranium was originally in the lowest stage of oxidation represented by UO_2. As the result of subsequent oxidation, whether by autoxidation as advocated by Ellsworth (1932, p. 89) or the oxidizing effects of the meteoric solutions, many uranium minerals show definite layers containing high percentages of UO_3.[27] In the form of this higher valence state, uranium is probably more susceptible to leaching action than lead. Consequently, altered uranium minerals may show misleadingly high lead age ratios.

Another possibility, that would result in quite the opposite effect, is the loss of intermediate members of the radioactive series from the mineral. For example, the radioactive gases radon, Rn, thoron, Tn, and actinon, An, might escape to some extent from the mineral by diffusion. If this should happen, the end-product lead would be deposited elsewhere. Two factors oppose this loss: (1) These gases are relatively short-lived[28], hence are only present in minute amounts at any instant and could not travel far without decaying, and (2) these gases are so heavy that their rates of diffusion are extremely small. However, it seemed worth while to test this possibility in at least one instance.

The Swedish kolm is the only material of high radioactive content that is precisely dated by geologic evidence; distinctive fossils prove it to be of latest Cambrian age.[29] Ironically enough, this unique material is particularly resistant to analysis. The lead age ratios observed average about 400 million years, whereas the lead isotope ratio for the kolm shows a value of 770 million years (Nier, 1939, p. 159). Since escape of radon would be in the direction to account for this difference, measurements were made to evaluate this factor. A 750-gram hand sample of kolm was

[26] Carbon-hydrogen and hydrocarbon minerals are found associated with uraninites and other radioactive minerals in pegmatites in Canada, Sweden and Russia (Ellsworth, 1932, p. 88).

[27] A method of estimating geologic age by the proportion of oxygen to that of uranium and lead, assuming that all the uranium was originally in the form of UO_2, was suggested by Ellsworth and has been the subject of investigation by Lane and Khlopin. This method, however, requires study of the crystal structure and density of the mineral, as well as accurate determinations of oxygen and the ratio of UO_2 to UO_3.

[28] $T_{Rn} = 3.82$ days, $T_{Tn} = 54.5$ secs., and $T_{An} = 3.92$ secs.

[29] For further details, including various analyses and geologic notes, *see* National Research Council, 1931, p. 299.

found to lose 0.25×10^{-9} curies of radon per 24 hours when sealed in nitrogen at atmospheric pressure. Gamma-ray measurements on the sample indicated a radium content of 0.8×10^{-6}g, which corresponds to about 0.3 per cent uranium. These results indicate an emanating power

TABLE 6.—*Lead age ratios of selected minerals*

Mineral	Locality	Geologic age	Age ratio in 10^6 years
Samarskite	Glastonbury, Conn	Pre-Triassic	270
Pitchblende	Jachymov, Bohemia	Late-Paleozoic	220
Thorite	Brevig, Norway	Permian (?)	230
Kolm	Güllhögen, Sweden	Latest Cambrian	400
Bröggerite	Karlhus, Raade, Norway	Pre-Cambrian	900
Cleveite	Aust-Agder, Arendal, Norway	Pre-Cambrian	1000
Uraninite	Keystone, S. Dakota	Pre-Cambrian	1500
Uraninite	Sinyaya, Pala, Carelia, Russia	Pre-Cambrian	1850

For geologic notes and references, *see* National Research Council, 1931, p. 7; Evans, *et al.*, 1939, p. 944; and Nier, 1939, p. 139.

of only 0.15 per cent, which is hardly sufficient to account for any significant loss of radiogenic lead from the mineral due to diffusion of radon.[30]

LEAD TIME SCALE

As we have seen in the introduction to this paper, the real benefit to geology of the radioactive methods of age determination results from the establishment of a quantitative time scale and its use for correlation purposes. In building up this time scale, the rocks or minerals measured must not only fulfill the four basic requirements but in addition must be of well-defined geologic age. Unfortunately, only a very limited number of samples that have been measured satisfy these criteria for fixed points in the time scale. Essentially all the helium age ratios, prior to the recent work on magnetites, must be discarded for the reasons given. Most of the early lead age ratios can be considered only as maxima because of the failure to consider any correction for ordinary lead. Many other lead age ratios are of little or no value, because the measurements were made on badly altered material. Of the remaining radioactive minerals that apparently satisfy the four basic requirements, only the few shown in Table 6 can be considered as being even approximately well dated geologically. With the exception of the kolm, the span of possible horizons in these specimens is so great that they can hardly represent anything approaching fixed points in the time scale. These results clearly point out

[30] The possibility remains that the meteoric waters might remove a much higher fraction of the equilibrium amount of radon.

the need for more detailed study of the geological evidence concerning the age of radioactive minerals. For the present, therefore, the lead time scale derives its strength from the fact that a larger number of age ratios fall in the proper geologic sequence although individually they cannot be specifically assigned to definite horizons. Most of the minerals listed under the lead method in Figure 10 have been arranged on this basis. Although these values present a rather impressive array, covering a broad range, they cannot be used for comparative purposes with helium age ratios for rocks except in a very general way.

If there were a clear-cut stratigraphic relationship between the geologic occurrence of a radioactive mineral, suitable for dating by the lead method, and a rock or mineral, suitable for helium age studies, an indirect but sharp comparison would be possible. Thus far these conditions have not been found. Instead, in general we are dependent upon separate geologic dating of the two materials. Hence, the comparison must be based upon the interdigitation of time scales containing numerous lead and helium age ratios covering a wide range of geologic time. It is the necessity for this type of comparison, plus the added complication that it is apparently quite difficult to assign geologic ages to radioactive minerals, that introduces so much uncertainty in the present status of radioactive age methods.

ISOTOPE RATIO METHODS

Because the two isotopes of uranium, UI and AcU, have different rates of decay, the proportions of the daughter elements and the end products, Pb^{207}/Pb^{206}, have varied systematically during geologic time. This variation can be measured in two quite different ways which provide two other methods of estimating geologic ages.

In discussing the lead method, the necessity for isotopic analysis of the separated lead was emphasized. Not only does such an analysis afford a measure of the amount of ordinary lead present, but it also provides a measure of the Pb^{207}/Pb^{206} ratio, which in turn gives an independent estimate of the geologic age of the specimen. Nier (1939, p. 159) has applied this method to a number of leads whose age ratios were also known from other data. These values are included in Figure 10. Among the 20 measurements by Nier on leads from 13 different uranium-bearing minerals, the Pb^{206}/U^{238} ages are in fair agreement with those previously calculated from Pb/U ratios. The Pb^{207}/Pb^{206} ages on eight of these minerals are in good agreement with the Pb^{206}/U^{238} ages, but serious discrepancies appear in the other five minerals. The differences are most likely due to alterations in the minerals not previously recognized as serious.

A distinct advantage of the Pb^{207}/Pb^{206} ratio method is that it is much less sensitive to alteration of the radioactive mineral than the Pb/U ratio. Nier (1939, p. 162) has considered at length the possible effects of alteration and concludes "that with the exception of isolated cases *any* simple alteration will manifest itself in a difference in the ages computed by the two different methods." Accordingly, where there is good agreement between these two independent methods on the same specimen, it is very convincing evidence of their mutual reliability.

The relative intensity of coloration of the Ac C and Ra C′ rings in certain pleochroic haloes provides the other, more approximate, method of determining the ratio of past actinium series activity to uranium series activity and hence the age of the mica in which these haloes are found. Artificial coloration studies (Henderson and Bateson, 1934, p. 563) show that the blackening of the pleochroic haloes is proportional to the alpha-ray ionization in the early stages of blackening. Subsequently, the blackening increases more and more slowly with increasing exposure to ionization. For these reasons care must be taken in the selection of haloes to be studied, and in general it is necessary to use minerals whose age is greater than about 400 million years. These restrictions limit the applicability of the method, and thus far only three micas have been studied quantitatively (Henderson, 1934, p. 591). Of these three specimens, only one, the Murray Bay biotite, has been dated by the lead method. The lead age ratio is 1070 million years, whereas Henderson obtained only 750 million years. However, the latter value was based on an actinium : uranium series activity ratio of 0.04, Nier's (1939, p. 150) more accurate value of 0.046 ± 0.001 brings Henderson's estimated age to 960 million years, in much better agreement with the lead value.

RUBIDIUM-STRONTIUM METHOD

Another method of geologic age determination has recently been suggested by Hahn and his coworkers (1937, p. 189). Ordinary rubidium is composed of two isotopes, Rb^{87} and Rb^{85}, with abundances of 28 and 72 per cent respectively (Nier, 1936, p. 1041). Only the heavier and less abundant isotope has been shown to be radioactive (Hemmendinger and Smythe, 1937, p. 1052). Rb^{87} is beta-ray active and decays, with a half period of about 10^{11} years, to stable Sr^{87}. In the application of the general age equation (1) to this method, the amount of radiogenic Sr^{87} is the numerator, while the denominator is directly proportional to the rubidium content of the material. Ordinary strontium also contains Sr^{87}, but in an abundance of only 7.5 per cent.

A handicap to the general applicability of this method is the rarity of rubidium. It occurs in small amounts in certain scarce lithium materials,

in microcline, and in rhodizite, a rare borate of Al, K, Cs, and Rb (Ellsworth, 1932, p. 12). The richest source of rubidium is lepidolite which in some cases contains as much as 3 per cent Rb_2O. An additional limitation is the quantitative determination of the minute amount of radiogenic strontium present, even in the oldest rubidium-bearing minerals. Hahn and his associates separated 147 mg of 99.7 per cent pure Sr^{87} (Mattauch, 1937, p. 189) from 1.011 kg of a lepidolite analyzed as containing 2.6 per cent Rb.[31]

At present the most serious uncertainty in the method is in the knowledge of the disintegration constant of rubidium. Evans (1940, p. 284-287) has recently reviewed the various measurements of $\lambda_{Rb^{87}}$ and selects 3.5×10^{-19} sec^{-1} as most nearly correct, although even this figure may be in error by 30 to 40 per cent. This value is based on Hahn's measurements on the lepidolite from Silver Leaf Mine, near Winnipeg, Canada (not from Great Bear Lake, as reported), assumed to be the same age (1800 million years from Pb/U) as the near-by Huron claim pegmatite (Ellsworth, 1932, p. 105). Thus the situation is one of circuitous relationship. Lead ages are used to estimate the decay constant of rubidium which in turn is used to measure other Rb-Sr ages. An accurate experimental determination of λ_{Rb}, while very desirable, is subject to large uncertainties owing to internal absorption and scattering of the soft beta-rays, $E_{max} = 0.13$ Mev (Libby and Lee, 1939, p. 251), within even relatively thin sources of rubidium.

The logical extension of the Rb-Sr method in the future is its application to a number of suitable minerals of widely varying geologic age, the interpretation of the results being one of over-all agreement with other age measurements.

POSSIBLE POTASSIUM METHODS

Potassium, like its heavier chemical analog, rubidium, is also beta radioactive. However, whereas no gamma rays have been observed for rubidium, potassium is known to emit a homogeneous gamma radiation of quantum energy 2 Mev, the ratio of the number of quanta to the number of disintegration electrons being about 3 : 100.

The beta-active isotope has been shown (Hemmendinger and Smythe, 1937, p. 178-182) to be K^{40}, the least abundant of the three main isotopes, K^{39}, K^{40}, and K^{41}, present in fractional abundances of 0.932, 0.00011, and 0.068 respectively (Nier, 1935, p. 283). Accordingly, with the emission of a negative beta-ray, K^{40} changes to Ca^{40}. Unlike rubidium, however,

[31] These values are from a personal communication from Professor O. Hahn to Professor A. C. Lane and are slightly different from those quoted by Hahn *et al.* (1937, p. 189). Since there is a possibility that the recovery of strontium was not complete, the 147 mg. represents a lower limit which Hahn thinks may be 10 per cent too small.

the Ca^{40} decay product of potassium is the predominantly abundant isotope (96.76 per cent) of ordinary calcium.

Because potassium is so generously distributed in terrestrial materials and has a long half life ($T_{K^{40}} = 1.06 \pm 0.3 \times 10^9$ years), it would be particularly suited for geologic age measurements. However, because ordinary calcium is commonly associated with potassium, it would be difficult to evaluate accurately the numerator of the general age equation as applied to the $K^{40} \rightarrow Ca^{40}$ transmutation. It is conceivable that certain potassic minerals might originally have been quite free of calcium. These minerals would serve as valuable indices of age, but accurate isotopic analyses of the quantitatively separated calcium would be even more essential in these studies than in lead age measurements.

Another possible age method based on the decay of potassium has been suggested by Evans (1940, p. 282), although no direct confirmation of this idea has yet appeared. It appears that the high energy, low intensity gamma radiation of potassium is not associated with the beta-ray disintegration of K^{40}. Instead it is tentatively considered to be due to K-electron capture by a definite fraction of the K^{40} nuclei. The decay product of this change would be A^{40}, the most abundant (99.63 per cent) isotope of argon.

Postulating this to be the process, only about 7×10^{-6} cc of A^{40} (referred to O° C and 760 mm Hg) would be formed in 100 million years. While it would be possible to determine such quantities of argon fairly accurately this method would be complicated by the likely presence of much larger amounts of ordinary argon, of nonradiogenic origin. Shepherd (1938, p. 311) has found argon to be a very common constituent of both lavas and plutonic rocks. He reports at least traces, which for his measurements means more than 10^{-4} cc per g, for 25 out of 29 rock samples. These concentrations are several thousand times greater than could have been produced by the potassium present, which ranges from 0.11 to 4.46 per cent. Thus, while both potassium age methods are worthy of further investigation, it does not appear likely that either of them will be capable of extensive application.

CONCLUSIONS

From a critical review of the fulfillment of the basic requirements by the various radioactive age methods, it is concluded that: (1) the retentivity of rocks for helium is the major source of uncertainty in the present application of the helium method, (2) analytical errors, earlier misconceptions, and the lack of attention to the basic requirements have necessitated the discarding of much of the age research of the past 3 decades, (3) isotopic abundance measurements are essential in the de-

termination of ages by the lead method, (4) more specific geological dating is needed in the formulation of a useful lead time scale, (5) more direct comparisons between the lead and helium age ratios are urgently needed, (6) although other methods have been discovered, the lead and helium methods still remain the outstandingly promising radioactive methods of quantitative age determination, and (7) recent developments in the application of the helium method to selected common rock minerals promises a method of dating igneous rocks that should be of wide application as an aid to geologic correlations.

GEOLOGIC NOTES ON ROCK SPECIMENS REFERRED TO IN TABLES

4A$_1$ Chelmsford granite collected by N. B. Keevil from H. E. Fletcher Co. quarry, N. Chelmsford, Mass. Age: possibly Mississippian (Emerson, 1917).

7A Rockport granite, Lat. 42° 40′ N., Long. 70° 35′ W., collected by N. B. Keevil from Upper Pigeon Hill Quarry, Cape Ann, Mass. Intrudes Salem gabbro diorite, which in turn cuts pre-Cambrian rocks and is possibly of Devonian age. It is cut by diabase dikes interbedded with Triassic sedimentary beds at Deerfield and Mt. Holyoke, Mass. Age: probably pre-Triassic and post-Cambrian.

8A Rockport xenolith, fine-grained and cognate, from the same large hand specimen as 7A.

11A Granite collected by N. B. Keevil from quarry in Franklin, Me., (*see* Dale, 1923, p. 219). Age: Middle Paleozoic.

12A$_1$ Quincy granite collected by N. B. Keevil from J. S. Swingle quarry, Quincy, Mass. Age: possibly Mississippian, probably late Paleozoic.

15A Fitchburg granite collected by N. B. Keevil from Rollstone Hill quarry, Fitchburg, Mass. Age: probably Carboniferous.

17A Granite collected by N. B. Keevil from Barryfield Quarries, 2 miles east of Kingston, Ont. Age: post-Grenville and pre-Keweenawan.

23A Rhyolite collected by A. D. Howard 100 feet north of Tower Creek Bridge, Yellowstone Park.

28A Obsidian collected by C. Max Bauer from Obsidian Cliff, Yellowstone Park.

31A Lebanon granite collected by E. P. Kaiser (1938, p. 107) near Hanover, N. H.

33A Pegmatite sample collected by A. C. Lane, 1937, from Ruggles Mine, Grafton, N. H.

1B Porphyritic basalt collected by A. D. Howard from southeast base of Crescent Hill, Yellowstone Park.

3B Crescent Hill Basalt collected by A. D. Howard ¼ mile west of crossing of Geode Creek, Yellowstone Park.

4B Basalt from the well-known Giant's Causeway in Ireland, submitted as a large hand specimen for intercheck measurements by Arthur Holmes (1937a, p. 155); *also see* 5I and 7I.

6B Basalt from Lat. 40° 40′ N., Long. 40° 45′ W., collected from a flow by A. C. Hawkins, 1936, 50 feet from the surface in a working quarry at Oldwick, N. J. The top of the flow has been removed by erosion, and the bottom is not visible. Similar flows in the area of slightly different periods of eruptivity have been identified as being of Triassic age from fossils in adjoining sedimentary beds (red shales and sandstones). Age: probably Triassic.

7B Basalt collected by A. D. Howard ¾ mile east of crossing of Geode Creek, Yellowstone Park.

10B Basalt collected by A. D. Howard from Overhanging Cliff, Yellowstone Park.

14B Diabase from Lat. 43° 15′ N., Long. 79° 2′ W., collected by P. Price 3 feet from chilled contact of a north-south dike at the 17th level (1975 feet) of the Horne Mine, Norando, Quebec. In a previous publication (Evans, *et al.*, 1939, p. 946) this rock was indicated as post-Cobalt in age. Subsequently, a personal communication from H. C. Cooke informs us that this dating is very uncertain, and the most that can be said is that this rock is post-Timiskaming (Cooke, *et al.*, 1931, p. 143).

22B Norite. Lat. 45° 1′ N., Long. 109° 25′ W., collected July 1937 by A. C. Lane from the metamorphosed Stillwater complex of Quad Creek, Mont., 1 mile north of state line. The Stillwater complex is bordered by a Cambrian conglomerate at an erosional contact and is cut by granite and by basic dikes. Age: probably early pre-Cambrian.

24B$_1$ Trap rock, Lat. 44° 56½′ N., Long. 109° 29½′ W., elev. 9800 feet, collected from a small working quarry by A. C. Lane, July 1937, 2 feet from the margin of a 100-foot dike at Long Lake, Wyoming. The dikes of this series are intrusive into the early pre-Cambrian complex (Stillwater series), and appear to be older than the middle Cambrian rocks of Beartooth Butte, but the exact relations are obliterated by overburden for a considerable distance. Age: possibly pre-Middle Cambrian and post early pre-Cambrian.

25B Trap rock, Lat. 44° 58′ N., Long. 109° 29′ W., elev. 10,600 feet, collected by A. C. Lane, July 1937, as part of Beartooth Range suite, from center of 25-foot dike, dip 45 degrees, strike north-northwest.

26B Monchiquite dike, Lat. 55° 45′ N., Long. 5° 3′ W., submitted by Arthur Holmes in 1936 from Riasg Buidhe on the Island of Colonsay, Scotland. Geologic relations nowhere provide evidence of age beyond the fact that it cuts formations regarded as Torridonian. Age: post-Devonian.

27B Monchiquite dike, same as 26B but from Kilchattan instead of Riasg Buidhe.

32B Andesite collected by K. G. Bell (1939, p. 10) from Dalhousie Mt. in Nova Scotia.

37B Tower Falls basalt collected by A. C. Lane, 1937, in Yellowstone Park at about 1.4 of Yellowstone Valley Conference (1937, p. 16), 134 feet vertically and 30 feet horizontally from the outcrop surface. Age: Recent.

38B Trap, Lat. 44° 56½′ N., Long. 109° 29½′ W., elev. 9800 feet, collected by A. C. Lane, as part of Beartooth Range suite, from altered glassy margin of Long Lake Dike by new road from Red Lodge to Yellowstone, near Long Lake, Mont., at about 35 of Yellowstone Valley Conference (1937, p. 11). Age: possibly latest pre-Cambrian.

43B Trap rock from Gogebic Range, Mich., collected by L. M. Scofield, Jan. 16, 1936. "Normal" from the 26th level of the Brotherton mine and cuts the upper phase of the Palms (Huronian).

4I Granodiorite from La Motte, Quebec, collection of R. A. Daly. Other measurements by Keevil (1938a, p. 145).

5I Tholeiite from Cleveland Dyke, Bolam County, Durham, Eng., submitted as a small hand specimen for intercheck measurements by Arthur Holmes (1937a, p. 118).

6I Hornblende granodiorite, Lat. 48° 14′ N., Long. 77° 54′ W., collected by W. C. Gussow in 1936 from a specimen freshly blasted from a road out 10 feet from the surface. The granodiorite is intrusive into a sedimentary complex of Keewatin or possibly earlier age and is thought to be older than the diabase dikes in the area and the Cobalt series to the southwest. Age: probably pre-Cobalt and post-Keewatin.

7I Quartz dolerite from Whin Sill, Scordale Beck, Westmoreland, England, submitted as a small hand specimen for intercheck measurements by Arthur Holmes (1937a, p. 118).

4M Hornblende gneiss obtained from collection of R. A. Daly, designation 1737. Metamorphosed pre-Cambrian, known as Shuswap gneiss from British Columbia.

5M Feldspar augen gneiss submitted by R. A. Daly, designation 1848.

6M Granodiorite from contact phase of Castle Peak stock, Long. 120° 50′ W., collected by R. A. Daly during survey of 49th parallel, designation 1442.

7M Schist collected by A. C. Lane, 1937, as part of Beartooth Range suite. Specimen is from a hornblende schist inclusion in the orthogneiss; some premetamorphic material in it. Location about 34 of Yellowstone Valley Conference (1937, p. 11).

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HELIUM RETENTION IN COMMON ROCK MINERALS

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ABSTRACT

In the helium method of age determination the age of the material to be tested is given by the ratio of the content of helium to the content of radioactive elements in the material. In an unaltered igneous rock, where presumably the minerals are of the same age, it would be expected that the distribution of helium in the various minerals should correspond with the distribution of the radioactive elements. In each mineral the ratio of helium to radioactivity should be the same. This was found to be not the case.

Pyroxene and feldspar were separated from six samples of Triassic diabase. Age measurements on each yielded mean age ratios of 103 million years for the pyroxene samples and 36 million years for the feldspar samples. Six samples of late Triassic magnetite from ore occurrences in West Virginia, Pennsylvania, and Nova Scotia all gave age ratios closely grouped about a mean value of 134 million years. The constant age ratio, despite widely varying contents of radium and thorium, suggests that the magnetite has retained most of its helium.

Additional evidence on the high helium retentivity of magnetite is shown by further work on magnetite specimens of various geological ages. These age ratios appear to show a sequence and spacing compatible with geological knowledge, and the results are in fair agreement with corresponding age ratios given by the lead method.

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INTRODUCTION

The present paper reports the progress made to date in a fundamental investigation of the retention of helium by rocks and rock minerals. This question constitutes the major uncertainty in the application of the helium method of geologic age determination. At the beginning of this research, in the fall of 1939, the following features characterized the status of the helium method:

(1) Higher helium age ratios were observed for rocks of greater geologic age, as a general rule, but there were many exceptions.
(2) Acidic rocks gave lower helium age ratios than basic rocks of the same geologic age.
(3) In general, the time scale based on helium age ratios was considerably less than that given by the lead age ratios of radioactive minerals.
(4) Anomalously high helium age ratios were observed for certain rock specimens.

Preliminary helium age measurements on late pre-Cambrian rocks from the Sudbury district, Ontario, had suggested that this region might offer an unusually good opportunity for helium retention studies. Accordingly, a suite of type rocks were obtained and their helium and radium contents measured. The results, together with the calculated maximum age ratios, are summarized in Table 1. It was immediately

TABLE 1.—*Age measurements on Sudbury rocks*

Sample No.	Rock specimen	Helium in 10^{-4} cc/g	Radium in 10^{-12} g/g	Maximum Age-ratio* in 10^6 years
29B	Sudbury gabbro	3.29	0.096	1050
$20B_1$	Norite	2.68	0.55	151
29A	Micropegmatite	2.97	1.01	91
9A	Murray granite	16.48	1.34	379
$10A_1$	Creighton granite	9.7	1.64	182
26A	Creighton aplite	6.86	3.13	67
$15B_3$	Olivine diabase	11.3	0.26	1320

* Specimens not analyzed for thorium. An average thorium content would reduce these age ratios by about one half.

apparent from the extreme randomicity of the results that these rocks involved too many uncertainties to be of value for a fundamental investigation. Instead of continuing the work on this region, it was decided that the problem would best be furthered by restriction to rocks which involved as few uncertainties (or variables) as possible. For this purpose specimens of igneous rock were sought which fulfilled as many of the following prerequisites as possible:

(1) No apparent metamorphism since time of formation.
(2) Conditions of origin not in doubt, preferably with crystallization under low pressure to reduce possible inclusion of primary helium.
(3) Bi-mineralic in composition and of sufficiently large grain size to facilitate mineral separation.
(4) Opportunity for lateral and vertical range in sampling.
(5) Geologic age known.
(6) Sufficient radioactivity for accurate analysis.
(7) Preferably glaciated surface to minimize effect of deep weathering.

The Triassic Palisade diabase and Watchung flows of New Jersey appeared to be satisfactory material, and large, 15 to 25 pound, specimens were collected from quarries and road cuts over a lateral range of 60 miles and up to 150 feet below the surface. Collection localities and petrographic notes for these specimens are included in a later section.

HELIUM LOSS

REGIONAL SCALE

Before beginning detailed work on helium retention, a study was made of the helium content in various parts of the Triassic rock masses to discover if there were any general relationship between helium content and depth of specimen below the surface, grain size, or proximity to the boundaries. Although the actual position of each specimen, with respect to the top or bottom of the flow or sill, was not closely known, it was assumed that a sufficient number of different positions were represented in the sampling to reveal any major variations existing. Of course, uneven distributions of radioactivity would cause similar uneven distributions of helium, and thus other minor variations would be masked. Hence, only large-scale differences were sought. Table 2 presents the results of these analyses. It will be noted that a remarkably uniform distribution of helium in each rock mass is indicated. From these results, it appears that the helium retention within the rock mass is independent of the location with respect to the surface or boundary.

CRUSHING AND GRINDING

As indicated by Goodman and Evans (1941, p. 517), conflicting observations have been made on the possible loss of helium during the preparation of the sample for measurement. Accordingly, a number of tests were made as a check on this possible source of error in the analytical technique.

Helium analyses were made on the same rock sample ground to sizes varying from ¼ inch to — 150 mesh, with the results shown in Table 3. Within the experimental uncertainty, there is no evidence of loss of helium from any of the samples over the lengths of time indicated.

TABLE 2.—*Distribution of helium in rock masses*

Sample No.	Rock specimen	Depth below surface in feet	Helium in 10^{-5} cc/g
	1st Watchung		
$9B_5$	Somerville	50–100	0.42
$9B_4$	Scotch Plains	70	0.45
$9B_3$	Scotch Plains	35	0.59
$9B_2$	Summit	30–50	0.61
	2nd Watchung		
$9B_1$	Murray Hill	25	0.75
$9B_6$	Livingstone	5	0.67
$9B_7$	Preakness Mtn.	4	0.72
$9B_8$	Patterson	100	0.77
	3rd Watchung		
$9B_9$	Riker Hill	4	0.74
$9B_{10}$	Mt. View	7	1.49
$9B_{11}$	Packanack Mtn.	15	1.31
	Palisade		
$16B_1$	Staten Island	8	0.91
$16B_2$	N. Bergen	20	0.96
$16B_3$	Guttenberg	150	0.93
$16B_4$	Edgewater	70	1.02
$16B_5$	Kingston	20	1.01
	Standard Diabase		
18B	Virginia	+10	1.02

The values in Table 3 are also indicative of the variation in multiple helium analyses. The particular specimen studied has the coarsest texture of any in the suite, and, as we shall see in the next section, shows the greatest difference in helium and radioactive content between its mineral components. Therefore, this specimen would be expected to show the maximum variation in the suite. The range of helium values observed for the samples of ¼ inch material is only slightly greater than for the — 48 + 100 mesh samples, which suggests that the sampling uncertainty is less than the analytical uncertainty for these quantities of helium. In both cases the individual measurements are in agreement within about 10 per cent of the mean value.

AGE MEASUREMENTS

LATERAL VARIATION

Measurements of helium, radium, and thorium made on rock specimens from the Palisade sill over a lateral distance of 45 miles show a remarkably

uniform distribution. Age ratios calculated from these measurements [1] are given in Table 4 and are somewhat higher than the value obtained by Keevil and Goodman (Evans, *et al.*, 1939, p. 942, Oldwick basalt) on one other specimen of New Jersey trap rock. While these recent determinations are definitely not in agreement with each other within the

TABLE 3.—*Tests for helium loss on grinding*

Staten Island diabase
($16B_1$)

Size of material	Sample weight (in grams)	Time* (in days)	Helium in 10^{-6} cc/g
−¼ inch	10	1	0.82
−¼ inch	5	34	0.95
−¼ inch	5	68	1.03
−48+100 mesh	2	3	0.92
−48+100 mesh	4	4	0.84
−48+100 mesh	6	16	0.99
−48+100 mesh	7	17	0.80
−100 mesh	5	1	0.91

* Elapsed time between grinding and analysis.

analytical accuracy of the measurements, there is no evidence of a systematic variation with lateral distribution, such as might be expected if any significant migration of helium occurred over a regional scale.

SEPARATE MINERALS

Radioactivity.—Before considering the age ratios obtained for separate minerals, mention should be made of the distribution of the radioactive elements in rocks. Waters [2] (1909, p. 677; 1910, p. 903) appears to have been the first to measure the radioactivity of mineral separates from common rocks. Although the gold leaf electroscope used by Waters was not sufficiently sensitive to detect any activity in the rock as a whole, his conclusions concerning certain mineral constituents have since been amply confirmed. The major activity was found to be concentrated in minute inclusions of zircon, allanite, and sphene, with somewhat less in magnetite, apatite, and rutile. The dimensions of the individual minerals, sufficiently active to be measured by Waters' instrument, were less than the ranges of most of the alpha rays. Hence, it is not surprising that these minerals were found to contain only a fraction of the helium

[1] The method of calculation is discussed by Goodman and Evans (1941, p. 507).

[2] In view of recent developments, it is particularly interesting to note that Waters, in the first paper, stated the following: "The object of the investigation was to find in what minerals the radioactivity of igneous rocks is most concentrated, and, if possible to separate them with a view to their use as material for finding the age of the rocks in which they occur."

content expected. It is regrettable that for this reason these interesting initial investigations were discontinued.

Other workers, notably Strutt (1909, p. 298), Fletcher (1911, p. 102), and Holmes and Paneth (1936, p. 385), have made extensive measurements of the helium, radium, and thorium contents of zircons. The

TABLE 4.—*Distribution of helium, radium, and thorium in the Palisade sill*

Sample No.	Location	Helium in 10^{-5} cc/g	Radium in 10^{-12} g/g	Thorium in 10^{-6} g/g	Age ratio in 10^6 years
$16B_1$	Staten Island.....	0.91	0.24	2.14	62
$16B_2$	North Bergen.....	0.96	0.19	2.37	72
$16B_3$	Guttenberg.....	0.93	0.16	2.34	75
$16B_4$	Edgewater.....	1.02	0.14	2.08	92
$16B_5$	Kingston.....	1.01	0.17	2.60	76

radioactive content varies widely; accessory zircon has in general high activity, while larger crystals in zirconium-rich rocks are often only feebly radioactive.

Evans and Williams (1935, p. 441) have summarized the limited number of measurements that had been made up to 1935 on other igneous rock minerals. They also report radium measurements on mineral separates of two Lassen dacites. Keevil (1938, p. 406) has subsequently studied the distribution of helium, radium, and thorium in the Quincy granite.

It may be concluded in general that, although there are individual exceptions, the radioactivity of biotite and other ferromagnesian minerals is higher than that of quartz and the feldspars. As is well-known to petrographers, these minerals usually contain most of the zircon, allanite, and sphene which may account for the higher radioactivity of the biotite and other ferromagnesions. There is considerable evidence that the radioactive elements also occur as individual atoms within the crystal structure of the essential rock minerals.

Distribution of helium.—As the radioactive elements undergo spontaneous disintegration, the alpha particles (helium nuclei) are emitted with sufficient energy to penetrate the surrounding mineral matter a distance of 40 micra or less. After expending their energy, the alpha particles become normal helium atoms and appear to lodge themselves in the crystal structure of the mineral they have penetrated. This process may be pictured as an involuntary solid solution of the helium by the mineral. Unless the helium atoms escape from the solid structure and accumulate in open spaces, nowhere will radiogenic helium be present as gaseous inclusions. Since most rock minerals have dimensions much

greater than 40 micra, the major portion of the helium produced within a particular mineral is, initially at least, contained within the boundaries of that mineral. It might be expected that the firmness with which the helium atoms are held by a crystal structure would differ according to the atomic arrangement in the mineral.

TABLE 5.—*Measurements on separated minerals*

	FELDSPAR			PYROXENE		
Sample No.	Helium in 10^{-6} cc/g	Radium in 10^{-12} g/g	Thorium in 10^{-6} g/g	Helium in 10^{-6} cc/g	Radium in 10^{-12} g/g	Thorium in 10^{-6} g/g
$16B_1$.....	0.25	0.23	2.24	1.57	0.23	2.48
$16B_2$.....	0.31	0.18	1.76	1.46	0.18	2.51
$16B_3$.....	0.57	0.13	1.44	1.29	0.19	2.72
$16B_4$.....	0.35	0.12	1.82	1.37	0.14	2.36
$16B_5$.....	0.30	0.11	1.81	1.46	0.18	3.49
18B.....	0.38	0.14	2.10	1.52	0.25	2.22
Average.....	0.36	0.15	1.86	1.45	0.19	2.63

If a particular mineral permits migration and loss of helium atoms from its structure, a lower age ratio would be obtained for this mineral than for an adjoining one with a higher helium retentivity. The observed age ratio of the rock containing these minerals would then be the weighted mean of the age ratios of the individual minerals and would depend upon the proportion and helium retentivity of all the minerals composing the rock. The true age of the rock would not be shown by direct analysis of the whole rock but could be found only from measurements on isolated minerals that have retained all their radiogenic helium.

Age measurements.—The present research has shown that the age ratio in feldspar is much lower than in pyroxene separated from the same rock. Six specimens of Triassic diabase were ground sufficiently to unlock most of the grains, and the minerals were separated in an electromagnetic separator. The final samples consisted of pure feldspar separates and almost pure pyroxene concentrates. The mineral compositions of the pyroxene concentrates were calculated from Rosiwal analyses of the original specimens.

The helium, radium, and thorium contents in the feldspar and in the pyroxene of each specimen of diabase are given in Table 5. The corresponding age ratios calculated from these value are compared in Table 6. The grouping of the age ratios for the two minerals clearly shows that there is a definite disparity.

The variation in the age ratios for the pyroxene is probably greater than would be the case if it could be separated from the feldspar more completely. Because of the extreme difference in helium age ratio, small quantities of feldspar would significantly affect the results obtained for the pyroxene fractions.

TABLE 6.—*Age ratios of separated minerals*

Sample No.	Age ratios of minerals in 10^6 years		Age ratios of rock in 10^6 years	
	Feldspar	Pyroxene	Calculated*	Observed†
$16B_1$	18	105	56	62
$16B_2$	28	109	82	72
$16B_3$	66	90	82	75
$16B_4$	37	119	93	92
$16B_5$	34	90	69	76
18B	35	102	92	98
Average	36	103	79	79

* Weighted mean from observed age ratios for separate minerals and Rosiwal analysis of their proportions.

† Observed on rock as a whole; *see* Table 4.

A comparison of age ratios for the rock as a whole is also included in Table 6. The mean value, calculated from the age ratios for the separate minerals and the Rosiwal analyses, in each case is in good agreement with that observed by direct measurement of the unseparated rock specimen. This correspondence furnishes additional evidence that there is no appreciable loss of helium upon crushing and grinding the rock sample.

DISCUSSION OF RESULTS

It seems probable that the age ratios shown by the feldspar, at least, are too low. If so, there are two reasonable ways in which such anomalous age ratios could have been caused; the feldspar has either lost helium or gained radioactivity during its geological life.

In thin section the feldspar is seen to be fresh with all evidence against its having undergone growth or recrystallization since the formation of the diabase. Bombardment of alpha particles out of the more radioactive into the less radioactive minerals cannot explain the anomaly because this effect would make the helium content of feldspar too high rather than too low. For the same reason, the loss of minute radioactive grains during grinding and separation does not afford an explanation.

There is a possibility that the whole rock has become uniformly contaminated late in its life by solutions containing uranium and thorium. At present this explanation does not seem likely, for unless there has been selective contamination of the feldspar no single quantity of uranium or thorium subtracted from the values observed for each mineral will bring the age ratios into agreement and at the same time keep them within reasonable limits for the Triassic age. It is also quite unlikely that contamination could be so uniform for the entire rock body.

The most plausible explanation, at present, is that a portion of the helium in some way has escaped from the feldspar. Keevil (1940, p. 311) has presented a theoretical consideration of the probability of migration of helium atoms held in the crystal structures of common rock minerals. He concludes that an appreciable helium loss over geologic periods of time is highly improbable. However, since helium leakage appears as the most likely explanation of the age ratios observed, it seems probable that Keevil's theoretical treatment fails to include all the factors involved.

Assuming that helium does escape from minerals, and in varying degrees for different minerals, it is possible that the fractional loss may be a fairly specific quantity for each mineral, even with widely varying helium concentrations. This state of affairs would afford an explanation of the grouping of the age ratios about the mean values observed for the two minerals in Table 6.

TRIASSIC MAGNETITES

Although the foregoing measurements indicated helium loss from feldspar, the evidence was inconclusive concerning the degree of retention of helium by the pyroxene. It seemed probable that information concerning this question could be obtained from age measurements on other Triassic minerals, since, if higher age ratios were observed for these minerals than for the pyroxene, helium loss from the latter would be suggested. For this purpose four specimens of magnetite from deposits associated with the Triassic intrusives in West Virginia and Pennsylvania, together with two specimens associated with Triassic flows in Nova Scotia, were obtained. The results of helium, radium, and thorium measurements and the calculated age ratios for these six magnetites are given in Table 7. The age ratios range in value from 126 to 145 million years with a mean of 134 million years. The six values are in good agreement within the analytical uncertainty of the individual measurements. Comparing these age ratios with those for the pyroxenes in Table 6, it is seen that the magnetite values are all larger than even the largest value of 119 million years observed for $16B_4$, and the mean value for the magnetites is about 30 per cent greater than the mean value for the

pyroxenes. Thus it appears likely that the pyroxene also has lost part of its helium but that the fractional loss from pyroxene is smaller than that from feldspar.

In addition to the numerous interchecking measurements discussed in the papers by Evans and Goodman, the agreement in the magnetite

TABLE 7.—*Age ratios of Triassic magnetite deposits*

Sample No.	Locality	Helium* in 10^{-4} cc/g	Radium* in 10^{-14} g/g	Thorium* in 10^{-7} g/g	Age ratio in 10^6 years
1	Cornwall, Pa.	22.4	31.6	2.44	126
2	Goose Creek, Va.	15.7	14.8	2.25	137
3	Boyertown, Pa.	17.6	27.8	1.32	133
4	French Creek, Pa.	8.6	8.5	1.07	145
5	Gerrish Mt., N. S.	7.8	4.9	15.4	128
6	Lakeville, N. S.	11.4	18.0	8.2	135

* The units used are smaller than in the preceding tables in order to make the variation between samples more apparent.

age ratios also affords evidence that the radium and thorium determinations are accurate on an absolute basis. The proportions of radium and thorium in these specimens varied over such a wide range that the age ratio for No. 5, the Gerrish Mt., N. S., specimen, is essentially a He/Th ratio while the age ratio for No. 6, the Lakeville, N. S., specimen, is essentially a He/Ra ratio, with intermediate variations for the other specimens. If any systematic error occurs in either the radium or thorium measurements, it should be reflected in a lack of agreement in the corresponding age ratios for these specimens.

MAGNETITES OF VARIOUS AGES

While the age ratios for Triassic minerals indicated that magnetite has a higher retentivity for helium than either feldspar or pyroxene, still it could not be concluded that magnetite retains all its helium. Accordingly, a new program of research was initiated to investigate what may be considered the specific retentivity of common minerals. As mentioned above, there is a possibility that this factor may be fairly constant for individual mineral species. As indicated by Goodman and Evans, no reliable helium time scale existed at the time the present researches were begun. Accordingly, as a tentative measure, it was decided to consider magnetite as having complete helium retentivity and to use it as a standard against which other minerals could be compared. For example, considering magnetite as unity, the above results indicate that the specific retentivity of feldspar is about 0.3 and that of pyroxene about 0.8. By testing other

mineral types in addition to further measurements on those already studied, it is hoped that some information may be gained on the absolute retentivity of helium in magnetites as well as the possible use of other minerals for age determinations.

TABLE 8.—*Age ratios of various magnetite deposits*

Sample No.	Specimen and geologic age	Helium in 10^{-6} cc/g	Radium in 10^{-12} g/g	Thorium in 10^{-6} g/g	Age ratio in 10^6 years
	Mid-Tertiary				
7	Great Western Mine, Utah	0.85	0.62	4.0	26
8	Black Magnetic Mine, Utah	0.98	0.67	1.4	37
	Early Tertiary				
9	Mémé, T. N., Haiti	0.72	0.33	0.31	60
10	Fierro, New Mexico	0.40	0.12	0.9	59
	Early Cretaceous				
11	Prince of Wales I., Alaska	3.53	1.23	0.64	81
12	Lynn Valley, Br. Columbia	1.26	0.42	0.44	88
	Late Jurassic				
	Texada I., B. C.				
13	M. I. T. specimen	1.66	0.34	0.50	130
14	Smithsonian Inst. specimen	2.35	0.51	0.22	131
	Late Triassic-Early Jurassic				
1	Cornwall, Pa	2.24	0.32	0.24	126
2	Goose Creek, Va	1.57	0.15	0.22	137
3	Boyertown, Pa	1.76	0.28	0.13	133
4	French Creek, Pa	0.86	0.085	0.11	145
5	Gerrish Mtn., N. S.	0.78	0.049	1.54	128
6	Lakeville, N. S.	1.14	0.18	0.08	135
	Carboniferous				
15	Magnitnaya, Urals	4.73	0.44	0.27	300
	Devonian				
16	Vysokaya, Urals	0.81	0.048	0.33	316
	Blagodat, Urals				
17	M. I. T. specimen	1.48	0.099	0.23	366
18	Smithsonian Inst. specimen	2.60	0.158	0.42	396
	Pre-Cambrian				
19	Kiruna, Sweden	64.9	0.300	16.2	1100
20	Hart Twp., Ont	26.6	0.600	0.51	1100

NOTE: The age ratios for samples 19 and 20 include a correction for the decay of uranium and thorium. (*See* Figure 6 of Goodman and Evans, p. 507.) The exact agreement is merely a coincidence.

The first step in this program has been the establishment of magnetite—helium age points in the geological time scale, against which the age ratios of other minerals may be compared. The results of this work to date are given in Table 8. The age ratios obtained range from 26 million years for a Mid-Tertiary magnetite to 1100 million years for two pre-Cambrian magnetites. Except for the two Late Jurassic specimens, for which the geologic evidence is somewhat uncertain, the age ratios are in the proper geologic sequence. Some evidence that magnetite retains most, if not all, of its helium is given by the fact that the magnetite-

helium time scale is in fairly good agreement with the post-Proterozoic points given by lead ratios and isotope ratios for radioactive minerals (Goodman and Evans, 1941, Fig. 10).

CONCLUSIONS

It is concluded from the investigations to date that:

(1) Age determinations by the helium method made directly on rock samples are likely to be in large error owing to loss of helium from some or all of the mineral constituents.

(2) Mineral species differ in their ability to retain helium. Feldspar loses a large proportion of its helium; pyroxene retains more of its helium than feldspar but less than magnetite.

(3) From the general agreement between the helium age ratios shown by magnetite and the existing time scale indicated by the lead method, magnetite appears to retain most, if not all, of its helium.

GEOLOGIC NOTES ON ROCK SPECIMENS REFERRED TO IN TABLES

29B Sudbury gabbro. 10 feet below original surface in road cut. Lot 3. Concession III, Graham Twp., Ontario.

$20B_1$ Sudbury norite. 15 feet below original surface in railway cut. Lot 1. Concession IV, Wisner Twp., Ontario.

29A Sudbury micropegmatite. 6 feet below original surface in railway cut. Lot 1. Concession III, Wisner Twp., Ontario.

9A Murray granite, Sudbury District. 5 feet below surface in railway cut. Lot 10, Range V. McKim Twp., Ontario.

$10A_1$ Creighton granite, Sudbury district. 5 feet below surface in road cut. Lot 9. Concession I, Snider Twp., Ontario.

26A Late aplite phase of the Creighton granite, Sudbury district. 4 feet below surface. Lot 1. Concession III, Snider Twp., Ontario.

$15B_3$ Olivine diabase, Sudbury district. 2 feet below surface. Lot 10. Concession V, McKim Twp., Ontario.

$9B_1$ Second Watchung basalt. Murray Hill, N. J. 25 feet below surface in quarry.

$9B_2$ First Watchung basalt. Summit, N. J. 30 to 50 feet below surface in quarry.

$9B_3$ First Watchung basalt. Scotch Plains, N. J. 35 feet below surface in quarry.

$9B_4$ First Watchung basalt. Same quarry as $9B_3$, 70 feet below surface.

$9B_5$ First Watchung basalt. Chimney Rock Quarry, north of Somerville, N. J. 50 to 100 feet below surface.

$9B_6$ Second Watchung basalt. Livingstone, N. J. 5 feet below surface in road cut.

$9B_7$ Second Watchung basalt. Preakness Mtn., N. J. 4 feet below surface in road cut.

$9B_9$ Third Watchung basalt. Riker Hill, N. J. 4 feet below surface in road cut.

$9B_{10}$ Third Watchung basalt. Mountain View, N. J. 7 feet below surface in road cut.

$9B_{11}$ Third Watchung basalt. Packanack Mtn., N. J. 15 feet below the surface in old quarry.

16B$_1$ Palisade diabase. Staten Island. 8 feet below surface in quarry.
Rosiwal analysis (per cent by volume): Feldspar 62
Pyroxene 36
Magnetite 1

Coarse-grained, fresh diabase. Feldspar entirely fresh, in labradorite laths with small amount of more acid intergrowth. The pyroxene occurs in large anhedral crystals, interstitial to the feldspar and commonly poikilitically including feldspar laths. Most of the pyroxene is fresh; where altered it has been replaced by a strongly pleochroic green biotite with a little chlorite and magnetite. Its composition is in a range between augite and pigeonite with an optic angle between 40 and 50 degrees.

16B$_2$ Palisade diabase. River Road, North Bergen, N. J. 20 feet below the surface in road cut.
Rosiwal analysis (per cent by volume): Feldspar 44
Pyroxene 53
Magnetite 3

Fine-grained diabase. All minerals are fresh. The pyroxene occurs in small rounded grains between the feldspar laths.

16B$_3$ Palisade diabase. River Road, Guttenberg, N. J. 150 feet below surface in quarry.
Rosiwal analysis (per cent by volume): Feldspar 49
Pyroxene 48
Magnetite 2-3

The rock has a fine, holocrystalline, diabasic texture with occasional large crystals of pyroxene. A small amount of acid feldspar occurs in graphic intergrowth between the labradorite laths. The pyroxene forms rounded and irregular grains. All minerals are fresh. There are two pyroxenes present, one with a composition in the range of pigeonite with an optic angle of about 15 degrees, and the other between pigeonite and augite.

16B$_4$ Palisade diabase. Edgewater, N. J. 70 feet below surface in road cut.
Rosiwal analysis (per cent by volume): Feldspar 47
Pyroxene 50
Magnetite 3

Fresh, fine-grained diabase. Feldspar laths show a minor amount of strain. The pyroxene is mostly pigeonite with optic angles close to 10 degrees.

16B$_5$ Palisade diabase. Kingston, N. J. 20 feet below surface in quarry.
Rosiwal analysis (per cent by volume): Feldspar 54
Pyroxene 45
Magnetite less than 1
Biotite less than 1

Medium-grained, fresh diabase. Feldspar mostly labradorite laths with some more acid intergrowths occurring interstitially. Pale-green pyroxene toward pigeonite in composition.

18B Standard Triassic diabase, Virginia. A number of rock samples have been prepared under the supervision of the National Bureau of Standards, for intercheck of radium and thorium measurements in various laboratories working on the radioactivity of terrestrial materials. One of these samples, the Triassic diabase, was used in the helium-age investigation.

1. Magnetite ore. Cornwall, Pa. Specimen of magnetite and maghemite with small amount of greenish gangue.

2. Magnetite in basic pegmatite. Goose Creek, Loudoun County, Va. Specimen of pegmatite with coarse, bladed augite crystals and basic feldspar. Considerable amount of coarse magnetite separated from it by electromagnet.
Age: Palisade. The pegmatite occurs in a diabase dike.

3. Magnetite ore. Boyertown, Pa. Specimen of fine-grained magnetite with small amount of greenish gangue.
Age: Palisade. Ore deposit associated with late Triassic diabase.

4. Magnetite ore. French Creek, Pa. Specimen of medium-grained magnetite with chloritic gangue and small amount of pyrite. Magnetite separated by electromagnet.
   Age: Palisade. Deposit associated with Triassic intrusives.

5. Magnetite. Gerrish Mountain, Nova Scotia. The specimen was taken from a magnetite-quartz veinlet in a Triassic flow.
   Age: Palisade.

6. Magnetite. Nova Scotia. 2 miles northwest of Lakeville. Associated with a Triassic flow.
   Age: Palisade.

7. Magnetite ore. Great Western Mine, Iron Springs, Iron Co., Utah. Smithsonian Institution Collection No. 42174.
   Age: Probably Early Miocene. Deposits associated with post-Eocene andesite which is similar to Miocene lavas of Wasatch Mtns. (Leith and Harder, 1908, p. 46.)

8. Magnetite ore. Black Magnetic Mine, Irontown, Utah. Smithsonian Institution Collection No. 42177.
   Age: Probably Early Miocene. *See* Specimen No. 7, Great Western Mine.

9. Magnetite and calcite. Germain Tunnel, Mémé T. N., Haiti. Specimen consists of large masses of foliated magnetite in a gangue of coarsely crystalline calcite. The magnetite is apparently pseudomorphic after hematite.
   Age: Tertiary. Deposits associated with intrusions cutting early Tertiary beds. (Geological Survey, Republic of Haiti, p. 443.)

10. Magnetite ore. Fierro, New Mexico. Specimen is unweathered but appears to have suffered from shear stress.
    Age: Early Tertiary(?). Deposit associated with porphyry belonging to a group of intrusive rocks placed as latest Cretaceous or, more probably, earliest Tertiary by Lindgren, Graton, and Gordon (1910, p. 35).

11. Magnetite in copper ore. Copper Mtn., Prince of Wales I., Alaska. Chalcopyrite and magnetite with gangue. Magnetite separated by electromagnet. Smithsonian Institution Collection No. 91562.
    Age: Early Cretaceous(?).

12. Magnetite ore. Lynn Valley, Vancouver, British Columbia. From Copper Queen Claim.
    Age: Probably Early Cretaceous or late Jurassic.

13. Magnetite. From the contact of the iron and copper deposits on Texada Island, British Columbia. The magnetite was separated from a greenish gangue by electromagnet. M. I. T. Collection No. 5943.
    Age: Probably Nevadan.

14. Magnetite. Texada Island, British Columbia. Same place and same age as Sample 13, but different collector. Smithsonian Institution Collection No. 42200.

15. Magnetite and hematite. Magnitnaya, Urals. Massive ore, somewhat oxidized. Magnetic, but with reddish color and streak.
    Age: Probably Upper Carboniferous or Permian. Associated with granitic intrusions cutting Lower Carboniferous strata (Zavaritsky, 1937, p. 92).

16. Magnetite ore. Vyssokaya, Urals. Magnetite separated from greenish gangue by electromagnet.
    Age: Devonian(?). Same age as Specimen No. 17, Blagodat, Urals.

17. Magnetite ore. Blagodat, Urals. Small amount of greenish gangue separated from magnetite.
    Age: Devonian(?). Deposit associated with syenite intrusion believed by Derwies (1937, p. 39) to be Middle Devonian.

18. Magnetite ore. Blagodat, Urals. Smithsonian Institution Collection No. 103318.
    Age: *See* Specimen 17.

19. Magnetite ore. Kiruna, Sweden. Fine-grained magnetite, separated from a gangue of apatite about 5 per cent by weight.
Age: Pre-Cambrian.

20. Magnetite ore. Carufel Copper Prospect, Hart Twp., Ontario. Fine-grained magnetite with a considerable amount of greenish gangue.
Age: The ore occurrence is believed to be of a contact metamorphic type associated with intrusive diabase similar to the dikes thought to be of Keweenawan age in the Sudbury district.

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COOLING OF THE EARTH

BY LOUIS B. SLICHTER

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ABSTRACT

In Part I observational data and hypotheses pertinent to the cooling problem of the earth are reviewed. The possible importance of heat transport by slow thermal convection currents is indicated. In Part II the theory of the cooling of a solid radioactive earth is presented by a simple method which identifies this problem with an equivalent one in the cooling of a nonradioactive earth. The facts as to slowness of cooling for temperature perturbations of long wave length are reviewed. The importance of the large heat capacity of the earth is emphasized by means of simple examples. If the earth is cooling the rate must be very small, but, if heating occurs, higher rates are possible. Thermal considerations indicate that solidification of the mantle probably occurred from the bottom up. Because the steady-state temperatures in a radioactive earth are significant in determining whether the earth is heating or cooling at depth, such temperature distributions have been computed for a dozen or more types of radioactive earths. It is probable that thermal equilibrium in the earth as a whole is far from achieved. The earth may be heating everywhere—cooling everywhere—or heating at some depths and cooling at others. The transient temperatures are expressed, and the transient heat flows computed for a number of types of suitable radioactive earths which are amenable to convenient computation. The case of heat flow in layered earths is especially examined. The observed surface heat flows fix definite upper limits to the total heat generation which may exist within about 100 or 200 kilometers of the surface. It is important to improve the reliability and scope of present information concerning (1) surface heat flows, (2) the structure and composition of the crust, and (3) the association of heat generation with crustal layers. Until better knowledge of all three of these factors in the same region is available, estimates of the amount of radioactivity existing in and below the crust will involve large uncertainties. Even granting excellent thermal information about the crust, it is possible to deduce by thermal theory only very limited information about regions below a depth of 200 or 300 km. Significant concentrations of radioactivity may exist all the way to the center without violating known criteria. At depth the problem of the cooling of the earth is nonunique to such a degree that little information about the deep interior may be deduced from purely thermal data.

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INTRODUCTION

Since Lord Kelvin's famous controversial estimate of the age of the earth in 1862, much attention has been given by geologists, physicists,

and mathematicians to the many aspects of the thermal problem of the earth. Jeffreys has published an excellent survey of modern developments in this subject in his book, *The Earth* (1929). The contributions of Ingersoll and Zobel (1913), Adams (1924), and Holmes (1933) are of much interest. Recently Spicer (1937) and Van Ostrand (1940) have computed and published tables relating to the cooling of homogeneous nonradioactive and radioactive earths. Lowan (1933) has contributed to the theory of the cooling of general types of nonhomogeneous radioactive earths, and Gutenberg (1939) has briefly summarized the present status of the subject in a chapter in his recent book. The limitations of space prevent the author's making adequate acknowledgment, or even reference, to the copious literature in this field which has developed since Kelvin's stimulating work. The appended bibliography is typical rather than comprehensive.

PART I. REVIEW OF HYPOTHESES AND OBSERVATIONAL DATA

INITIAL STATE OF THE EARTH

No accepted theory of origin.—It is usually postulated that the earth had a fiery beginning—more specifically that it was formed by the collision or near collision of a passing star (or two stars) with the sun. For many years attempts have been made to prove that such a cataclysm could actually account for the major features of the present solar system. However, these efforts have never been altogether successful, and recent work by Spitzer (1939) has revealed seemingly fatal objections to all solar collision theories of the origin of our planetary system. Irrespective of theories of origin (and without complete justification), it is generally held that the earth was originally hot and entirely in the molten state. If so, it may be established (*see* for example Jeffreys, 1929, p. 79) that a crust would cool to the point of solidification within a few tens of thousands of years, and that soon thereafter would have cooled sufficiently for an ocean to condense.

Constancy of surface temperature.—As noted by Kelvin, Jeffreys, and others the surface temperature would quickly reach an equilibrium essentially established by a balance between solar radiation and that reradiated to space—the heat conducted to the surface from below was always entirely too small to affect appreciably the surface temperature.[1] Assuming constancy of the sun's heat this mean temperature would not differ appreciably from that observed today. Furthermore it is well

[1] Today solar radiation supplies an average of about 4.2×10^{-3} cal./cm^{2}sec. to the surface. Conduction from below, probably about 1.2×10^{-6} cal./cm^{2}sec.

established from independent geological evidence that the mean surface temperature of the earth has remained remarkably constant[2] as far back in time as the record can be traced—namely, for more than half the earth's lifetime. This essentially constant surface temperature is conveniently adopted as the zero of reference.

POSTULATE OF A RIGID MANTLE

Three major divisions in the earth are recognized by geophysicists: The heavy *core* (mean density, 10.7) has a radius of about 3400 kms., or slightly over half that of the earth. It appears to be fluid and it is sometimes assumed to be composed of nickel-iron. Overlying the core is the thick lighter *mantle* (density, 3.4 to 5.7) generally held to have the approximate composition of ultrabasic rocks, but becoming richer in iron toward the core. Above the mantle is the thin *crust* consisting of several layers of lighter rocks whose total thickness is perhaps 20 to 60 kms. The crust appears to be subject to regional variations, and its structure differs especially between continental areas and the basin of the Pacific Ocean. There are minor structural features of interest in both the crust and the mantle, but the two major discontinuities in the earth occur between the crust and the mantle and between the mantle and the core. The seismological evidence supports conclusions based upon the tidal yielding of the solid earth in assigning to the mantle a coefficient of rigidity somewhat greater than that of steel. But the stresses involved in these seismic and tidal phenomena are of very short period relative to geologic time intervals, and it is not yet clearly established what effective rigidity the mantle may possess toward stress differences prolonged in geological time. However, it will be assumed in the thermal discussions which follow that the mantle, or at least the major part of it, is rigid, and has been rigid since its formation—"rigid" here meaning that convection velocities have been small in the sense of a paragraph which follows, so that conductive heat transport predominates. It is also assumed that the rigid mantle was formed so rapidly that its initial temperature was essentially independent of the distribution of radioactivity. In the absence today of any accepted theory of the origin of the earth it is perhaps futile to inquire further into the genesis of the primeval rigid mantle and fluid core here postulated. Nevertheless, because of the prevalence of the assumption that the earth was originally thoroughly molten, it is perhaps of interest to recall here a process by which the postulated earth might have been derived from a previously molten state.

[2] From the present point of view, variations in surface temperature of several tens of degrees are insignificant, since we are dealing with internal temperatures measured in thousands of degrees.

SOLIDIFICATION OF THE MANTLE

The different possible modes of solidification of an originally molten earth were well discussed by Hopkins (1839) over 100 years ago. In 1890 Kelvin outlined simple thermodynamical principles which are basic to modern discussions of the solidification process, such as those of Adams (1924) and Jeffreys (1929), but Kelvin lacked requisite thermal data about rocks upon which to develop his projected conclusions, and Adams appears to have first discussed upon a quantitative basis the process of solidification at shallow depths.

A thoroughly molten initial stage of the earth is here assumed to have existed, with temperatures sufficiently high to permit a free and rapid convection. In this stage, differentiation by density is presumed to have occurred to such an extent that the present heavy, molten core separated from the lighter material of the mantle, with the formation of a major convection system in the mantle and possibly another one in the core.[3]

Cooling in the mantle was essentially by convection, and accordingly the temperature distribution there was almost the adiabatic distribution. As the mantle rapidly cooled by convection solidification commenced at the depth at which the curve representing the adiabatic temperature first intersected, or became tangent to, the curve representing the depth-freezing point temperature. The adiabatic temperature gradient in the crustal magma is determined primarily by the ratio of the volume coefficient of thermal expansion to the specific heat in accordance with the equation (Adams, 1924):

$$\frac{\partial V}{\partial t} = Vg\alpha/C_p \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

The value of the gravitational acceleration g remains nearly constant throughout the mantle. Near the surface the absolute temperature V may be taken as about 1200°, the specific heat, C_p, as .25 cal./deg.gr., or nearly 1×10^7 ergs/deg.gr., and the thermal expansion, α, as 4×10^{-5}. With these values, the adiabatic gradient is about 4.6×10^{-6}° C./cm. The behavior of the adiabatic gradient at greater depths is very uncertain of estimate. Bridgman (1931, p. 136) states that the thermal expansion for liquids (not, however, silicates) at 12,000 atmospheres is generally between ¼ and ⅛ its value at atmospheric pressure. Similarly, the value of C_p in liquids generally first decreases slightly with pressure, then increases until at 12,000 atmospheres it has nearly recovered its original value; as a function of temperature the specific heat increases for nearly all rock minerals from a value of .2± .02 at 100° C. to about .3± .03 at 1000°C. Because of the decrease of the thermal expansion and the in-

[3] It is immaterial for present purposes whether convection existed in the core, or not

crease of the specific heat, it is probable that the adiabatic gradient decreases at depth.

The form of the depth-freezing curve is also difficult of estimation. The early work of Barus (1892) on basalt leads to estimates, based upon the Clausius-Clapeyron relation for the change of freezing point with pressure, of a freezing point gradient about tenfold greater than the adiabatic, or 3×10^{-5}°C./cm. This, if true, means that freezing first commenced at depth. Barus' work, however, was done on anhydrous samples which are certainly not representative of crustal magmas. Furthermore, the simple form of the Clapeyron relation does not apply for multiple component systems. The recent work of Goranson (1938) on the equilibrium of silicate-water systems under the temperatures of 800°-1200°C. and pressures up to 4000 bars (corresponding to depths of somewhat over 10 kms. in the earth) well indicates the complexity of equilibria relationships, even in the simple systems studied. In his experiments, the freezing point temperatures decreased instead of increased with increasing pressure throughout the observed pressure range, 0-4000 bars, but, as Goranson points out, the behavior at higher pressures of the parameters in the generalized Clapeyron equation which applies is such as to reverse the trend and lead to eventual increase of freezing point with pressure. Bridgman (1931, p. 206) in commenting upon the effect of pressure upon melting point says,

> ". . we have to take the present experimental evidence as making it exceedingly probable that the melting curves of all substances rise indefinitely, at a continuously decreasing rate, until pressures are reached so high as to introduce entirely new phenomena . . ."

There exists, unfortunately, little experimental evidence on this subject about silicates or magmas. Knowledge of equilibria in silicate systems is still confined to relatively simple systems, at relatively low pressures and temperatures.

Jeffreys (1932) revised his suggestion (1929) that solidification in the mantle commenced at a depth of several hundred kilometers, and supported the hypothesis that the process commenced at the outer boundary of the core itself. Such an assumption follows naturally if one accepts the hypothesis that the mantle is today predominantly solidified and rigid. Otherwise the cooling of the mantle to its present degree of rigidity from the postulated molten state would present difficulties. This subject will be further discussed in Part II. If solidification in the mantle proceeded from the bottom up, the magma would freeze at a relatively rapid rate, leaving the core essentially at its original temperature, and insulated by a rapidly thickening shell of silicates. During the process the surface of the earth would remain molten and would dissipate large amounts of

heat by radiation. Both gravitational differentiation and differentiation by selective freezing would be effective during the process. It is estimated that by such a process the complete mantle with its initial temperatures essentially at the freezing point would have formed in less than 20,000 years. The heat loss thenceforth would be by conduction and at a rate vanishingly small relative to that of the earlier convective stage. As a consequence of the rapid freezing the initial temperatures in the mantle would in no way be expressive of the equilibrium temperatures finally to be established by the radioactivity trapped in the rocks. At depth in the mantle the subsequent course of the temperature might be either increasing or decreasing as controlled by the concentration of the radioactive heat sources. Evidently, much freedom is permitted for further hypotheses concerning the subsequent thermal and mechanical history of such a mantle.

The outlined process of solidification involves selective freezing and possibly also some segregation by density. It is, therefore, probably not a reversible process, and any subsequent remelting of the solidified mantle would require higher temperatures—how much higher is, of course, unknown. Thus remelting of the mantle by radioactive heat would require heating through the necessary temperature increment, plus supplying the latent heat of fusion, which for rocks under ordinary conditions is approximately 100 cal./gr.

The heavier core was well insulated thermally by an enormous layer of relatively poorly conducting material. If originally liquid, it no doubt has remained liquid to this day and has probably maintained a nearly uniform temperature by convection or conduction or both. Its present temperature probably differs from its original temperature essentially by the amount of the radioactive heat which may have been produced in it.

As the mantle formed, its augmenting thickness would increasingly favor a tendency for the initiation of slow convection currents in the growing shell. Their possible contribution to the processes of heat transfer in the earth is important and will be considered in a following paragraph. However, the existence of such currents is still a question for research. If they exist, present theories for the cooling of the earth, which have all been based upon the hypothesis of a solid earth, will need radical revision.

RADIOACTIVITY OF ROCKS

It has long been recognized that the radioactive heat being generated in rocks is large enough to be a major influence in the earth's thermal history. The problem of estimating the amount of radioactivity which may exist at depths of several hundred kilometers is still little understood, but the approximate determination of the average radioactive con-

tent of the bulk of the crust seems within the scope of present possibilities. A beginning in this problem is just now being made. During the last several years an extensive and informative program for the study of radioactivity of rocks has been undertaken by Evans and his associates.

TABLE 1.—*Radioactive heat generation*

IN ROCKS

(After Evans and Goodman, 1941)

Rock type	No. of areas sampled	Density assigned	Heat generated	
			in 10^{-6} cal/g. yr.	in 10^{-13} cal/cc. sec.
Sedimentary	28	2.6	1.7±.6	1.4±.5
Acid igneous	43	2.7	5.0±.6	4.3±.5
Intermediate igneous	7	3.0	2.0±.4	1.9±.4
Basic igneous	54*	3.1	1.6±.1	1.6±.1
Deccan traps	1	2.98	.8±.13	.76±.12
Columbia River basalt	1	2.90	1.3±.08	1.2±.08

IN METEORITES

(After Paneth and Koeck, 1931, and Evans, 1938)

Type of meteorite	No. of samples	Density assigned	Heat generated	
			in 10^{-6} cal/g. yr.	in 10^{-13} cal/cc. sec.
Stony meteorites	several dozen	5†	.35	.52
Iron meteorites	several dozen	10†	.08	.25

* Includes the Deccan trap and Columbia River basalt samples.

† High densities are assigned to correspond with the densities appropriate at depth in the earth, in accordance with Washington's Model.

A summary of the results of Evans and Goodman (1941) in the form of mean values for the heat generation in the major rock types appears in Table 1. These averages are based chiefly upon their own work, with minor modifications due to their inclusion of selected determinations of others. Historically, the average amounts of radioactivity reported have tended to diminish as the subject has developed and techniques have improved. It is important to note that the present values of Evans and

Goodman are only about *one-third* those assigned to corresponding rock types 10 or 15 years ago.

Radium determinations have also been made on three or four dozen different stony meteorites and upon a like number of iron meteorites. The values for the radioactive heat generation in meteorites included in the table are mainly dependent upon the work of Paneth and Koech (1931) with some weighting of earlier work as summarized by Evans (1938).

Of the rock samples measured those which are probably most representative of the bulk of the material lying at depths of tens of kilometers are the Deccan traps and Columbia River basalts. Undoubtedly this basic basaltic material as now measured for radioactivity has lost a large part of its volatile constituents and probably a part of its uranium and thorium as it flowed into the lower-pressure and low-temperature environment (Hurley, personal communication). Therefore, we may expect the mean value of about 1×10^{-13} for these basalts to be somewhat exceeded by the source magmas for these rocks.

The average value of the heat generation in the random group of 43 acid rock samples in Table 1 was not expected to be applicable to the granitic layer of the crust. The composition as well as the thickness of this layer appear to be subject to important regional variations. (This inference receives additional support from recent detailed seismic studies of the crust in New England and in Wisconsin, by means of quarry blasts.) It seems probable that the group of granitic samples selected for laboratory determinations tends to represent a more highly differentiated type of rock than that representing the bulk of the granitic layer of seismology. Thus, if the seven granites representing the deeply eroded pre-Cambrian Canadian Shield be selected from the 43 acid rock specimens in the table it is found that their mean value of heat generation is only ⅓ that of the general mean—that is to say, it is quite comparable with the lower values listed for basic rocks. This suggests an average value for the granitic layer considerably lower than the value 4.3×10^{-13} listed. The measurements of Evans and Goodman indicate that the selected samples of both basic and acid rocks which are possibly most representative of the large bulk of crustal materials are notably lower in radioactivity than the listed general averages. Such preferred samples indicate that the heat generation in the granitic layer may be as low as 1 or 2×10^{-13} cal./cc.sec., and in the basaltic layer 1×10^{-13}. Much remains to be done in establishing the average composition of the crustal layers and in determining the probable mean radioactivity of both the granitic and basaltic layers.

RADIOACTIVITY AT DEPTH

It is generally held that the radioactivity must be essentially restricted to the upper few tens of kilometers of the crust and must diminish in concentration very rapidly with depth. Thus Jeffreys (1929, p. 144) writes, "No satisfactory alternative to this hypothesis is known, while several lines of evidence support it." Also, in Holmes' words, (1933, p. 171), "We must conclude with Strutt (Lord Rayleigh) that the radioactive elements are practically confined to a radioactive layer which corresponds with a crust a few tens of kilometers in thickness." And (1915, p. 64) ". . . the view that the earth is getting hotter can not be accepted." Bowen (1928), however, takes the opposite point of view. "The plain facts are that there is nothing that can be called definite knowledge to preclude the possibility that the earth is heating up." He also states in regard to the hypothesis of a rapid dimunition of radioactive content with depth,

"This result is not acceptable. Below the granite layer there must, in any rational scheme of things, be intermediate and basic rocks whose radioactive content is far from negligible. Below these again the ultrabasic rocks must be regarded as having the lower but still important quantity of radioactive substances usually found in ultrabasic rocks."

There is geochemical support to the inference that radioactivity has been concentrated in the upper layers.[4] However, the *degree* to which upward concentration of radioactivity has actually been accomplished may not well be estimated. No purely thermal criteria appear capable of assigning any but wide tolerances for the amount of radioactivity that may exist at depth. Indeed, the heat generation at depth may be comparable with that observed in stony or iron meteorites, or even with that inferred for deep crustal basalts, without leading to excessive heat flow at the surface. Accordingly, it is believed that no cogent thermal reasons exist for rejecting the hypothesis of a heating earth, or for concluding that an extreme degree of concentration of radium has occurred at the surface. Whether the tendency for heat generation to weaken or melt the mantle can be used to determine approximate upper limits for the amount of radioactivity at depth is a question still full of speculative possibilities.

[4] R. W. Goranson (personal communication) writes, "We know that thorium and uranium will rob iron of its oxygen. Thus they must lie in the oxide layer. We know, furthermore, that they tend to concentrate in the oxygen-rich rather than oxygen-poor oxides and in fact are found most abundantly in the very latest magmatic concentrates.

"It is to be expected then that the radioactive material of the earth will be concentrated in the upper layers of the earth. The evidence obtained from radioactive measurements on rocks corroborate such a picture. This latter evidence has little per se importance because of crustal contamination of the magma and thus of the rocks available to us."

AGE OF THE EARTH

Radioactive determinations of the age of old crustal rocks indicate that the time which has elapsed since solidification of the oldest rocks is about 1½ to 2 billion years. With the mode of formation of the crust which has been described, this also represents the interval elapsed since

TABLE 2.—*Thermal conductivity of some igneous rocks*

(After Birch and Clark, 1940)

Rock type	Assigned density	No. of regions represented	Conductivity, in 10^{-6} cal/cm. sec. deg. C			
			At 50°C		At 200°C	
			Range	Mean	Range	Mean
Granite.....	2.67	4	5.6–7.8	6.7	5.1–6.5	6.0
Anorthosite....	2.73	3	4.0–4.5	4.2	4.3–4.7	4.4
Diabase.....	2.98	3	5.0–5.5	5.2	5.0–5.4	5.1
Gabbro.....	2.98	3	4.7–5.4	4.9	4.7–5.1	4.9
Pyroxenite.....	3.23	2	9.2–10.0	9.6	7.8–8.7	8.3
Dunite.....	3.3	3	10.–11.4	10.5	7.5–8.8	8.1

the mantle was essentially at its freezing point temperature throughout. We shall adopt, then, as the initial time for the thermal problem one about 1½ to 2 billion years ago, when the mantle is assumed to have solidified.

THERMAL CONDUCTIVITY AND DIFFUSIVITY OF ROCKS

Birch and Clark (1940) have recently published the results of their excellent measurements concerning the thermal conductivity of rocks and minerals in the temperature range 0°- 300° C. A digest of their values for certain common igneous rocks is furnished in Table 2. At the higher part of the observed temperature range, the conductivities tend to converge, and at 200° C. all samples, igneous or sedimentary, measured by Birch and Clark have values between .004 and .009 cal./cm.sec.deg. Thus, the possible range for the thermal conductivity appears to be narrowly limited, if we may judge from surface results. But methods for extrapolation to conditions of far higher temperature and pressure are not yet known, and it is uncertain what the conductivity may be at depth.

The thermal diffusivity is obtained by dividing the conductivity by the density and the specific heat. At a given temperature the specific heats of nearly all igneous rocks are nearly the same; for example,

0.20 ± 0.02 at 100° C.; but the values all increase with temperature and, at 1000° C., lie in the range 0.3 ± 0.03. Values of the density of the common igneous rocks of Table 2 are listed in column 2 of the table. The range of density of these rocks is small. At depth (Bullen, 1940) the density of the earth is reasonably well known. It varies between 3.4 to 5.7 in the mantle; in the core, the mean density is between 10.6 and 10.8. The uncertainties in the values of the diffusivity at depth reside, therefore, chiefly in uncertainties regarding the conductivity and specific heat. Relative to the thermal conductivity, the diffusivity appears to decrease with depth.

OBSERVED HEAT FLUX IN CONTINENTAL AREAS

An evaluation of the heat flow to the surface from below requires both the measurement of the geothermal temperature gradient and the thermal conductivity of the rock. Although, as it has been noted, the conductivities of different igneous rock types are much alike, the sedimentary rocks vary as to conductivity throughout a wider range. Most of the numerous measurements of geothermal gradients have been made in sedimentary rocks, under conditions where the conductivity of the rock was not well known. The few exceptions are the excellent recent measurements by Bullard (1939) in South Africa, Benfield (1939) in England, and the comprehensive studies by Hotchkiss and Ingersoll (1934) in the deep mine of the Calumet and Hecla Copper Company in the Michigan Peninsula.[5]

Table 3 lists the heat flow results of Ingersoll (1934), Bullard (1939), and Benfield (1940).

By "equilibrium heat flow" is meant that which would occur in a crust undisturbed by recent surface-temperature changes, such as those occasioned by the retreat of recent glaciation. The heat flow represents both the heat liberation by shallow and (to a minor extent) deep radioactivity and that escaping from the earth's original store of heat.

Much more exhaustive heat-flow studies, with careful attention to correlations with the other pertinent geophysical factors, are needed. Continental, insular, and, insofar as possible, suboceanic areas need to be investigated and compared.[6] It is commonly believed that the local heat flux is predominantly occasioned by relatively shallow distributions

[5] Ingersoll measured the diffusivity, rather than the conductivity, of the Calumet and Hecla trap rock, obtaining a value of .0075. With a density of 2.8, and a specific heat of .22, this corresponds to a conductivity of .0046. Birch and Clark's measurements upon similar igneous rocks indicate, however, a somewhat higher value, .0052; and this value is adopted, pending results of measurements upon the rock itself, which it is expected will become available soon. The equilibrium geothermal gradient was 1.96×10^{-4} °C./cm.

[6] Suboceanic areas appear to differ structurally from continental areas only at shallow depths. Below the crust, results of seismology indicate that the earth has a high degree of symmetry. If this be true, it is to be expected that the radioactivity in sub-oceanic areas will differ from that in continental

of radioactivity. It is important to learn what degree of correlation exists, in areas where the crustal structure has been well established by seismic methods, between the observed heat flow and the carefully estimated regional crustal heat generation.

TABLE 3.—*Observed heat flow in crust*

Location	Equilibrium heat flow in 10^{-6} cal./cm²sec.	Average
Michigan		
Calumet and Hecla	1.0	1.0
South Africa		
Jacoba	0.95	
Doornhoutrivier	0.97	
Gerhardminnebron	1.28	
Doornkloof	1.20	
Reef-Nigel	1.03	
Dubbeldevlei*	1.52	1.16±0.09
Britain		
Balfour	.99	
Holford	1.04	
Blythswood	1.81	
South Balgray	2.23	
Hankham	1.03	1.42±0.14
	Average	1.20×10^{-6}

* This location about 500 miles southwest of the others.

The author has reviewed a considerable amount of data concerning observed geothermal gradients and has assigned, insofar as possible, rock conductivities suitable to the geological areas involved. These data include bore-hole gradients in the Canadian Shield (communicated by the courtesy of L. C. Graton), results in 6 additional North American areas, 1 South African district, and 18 European borings. The mean heat flux so computed was 1.15×10^{-6}. The method is not ideal, and the result is of uncertain accuracy; but the sample taken was fairly large, and the result tends to support the view that the mean heat flow is not far

regions only at shallow depth. Accordingly, the difference in heat flow between continental and oceanic areas is probably not a fundamental factor in the cooling of the earth as a whole. Since the observational data for the problem are almost entirely obtained in continental areas, the thermal problem here being considered is really equivalent to the cooling of an earth of purely continental type. But an earth of oceanic type would differ only by the absence of 10 or 20 kms. of overlying granitic material in the outer crust. This is not a major change.

from 1.2×10^{-6} cal./cm²sec. The subject is little explored, however, and regional values in the range 0.6×10^{-6} to 2.5×10^{-6} seem entirely possible. It would be most desirable to reduce this range of uncertainty through further field studies.

COMPARISON OF TRANSPORT OF HEAT BY CONDUCTION AND CONVECTION

The magnitude of the heat which could be transported by even exceedingly slow convection currents in the mantle of the earth (if these exist) is great compared to that which can be transported by conduction. In the following, the question of the relative effectiveness of these two processes in the interior of the earth is examined, and the order of magnitude of the convection velocities which would be thermally significant is established. Consider a sphere of uniform conductivity, in which convection is occurring only in meridional planes—that is, with velocity components v_r and v_θ, the longitudinal component v_ϕ being zero. Transport of heat under these circumstances is expressed by the equation

$$\rho c \frac{\partial T}{\partial t} = \sigma \left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r} \frac{\partial T}{\partial r} \right) - \rho c \left(v_r \frac{\partial T}{\partial r} + \frac{v_\theta}{r} \frac{\partial T}{\partial \theta} \right) \quad \ldots\ldots\ldots\ldots\ldots\ldots (2)$$

Here T denotes the temperature; t the time, ρ the density, and c the specific heat. At the center, $r = 0$, $\frac{\partial T}{\partial r}$ must vanish. Thus $\frac{\partial T}{\partial r} = \frac{2}{r} \int_0^r \frac{\partial^2 T}{\partial r^2} dr$, namely twice the mean value of $\frac{\partial^2 T}{\partial r^2}$ on the interval $(0,r)$.

Now the familiar types of temperature distributions which have been postulated in the earth agree in letting the gradient dT/dr decrease gradually in absolute value from the surface to the center, where it vanishes—that is, $\frac{\partial^2 T}{\partial r^2}$ is uniform as to sign. Under such circumstances the average value, $\overline{\frac{\partial^2 T}{\partial r^2}}$, has a magnitude comparable to the local value $\frac{\partial^2 T}{\partial r^2}$; and twice this average value (which is the same as $\frac{2}{r} dT/dr$) will nowhere be greatly less than the local value. This inference may be easily verified by examination of the temperature distributions in the earth which have been proposed in the literature, such as those of Adams (1924), Jeffreys (1929), and Gutenberg (1939). In the earth the radial temperature gradient $\frac{\partial T}{\partial r}$ in general exceeds the transverse gradient $\frac{\partial T}{r \partial \theta}$, whereas by virtue of the condition of continuity of the flow the maximum values of the components of velocity v_r and v_θ are necessarily of approximately the same magnitude. Thus the convective term $\rho c v_r \frac{\partial T}{\partial r}$ represents the greater part of the convective transport of heat, whereas the term $\sigma \frac{2}{r} \frac{\partial T}{\partial r}$ represents the order of magnitude of the conductive transport. It is clear,

therefore, that the convective transport will predominate whenever $\rho c v_r$ greatly exceeds $\frac{2\sigma}{r}$; namely when

$$v_r >> 2k/r \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (3)$$

where k is the thermal diffusivity, $\sigma/\rho c$. If surface measurements offer any guidance, the diffusivity at depth will not be large—perhaps of the order .01. Thus when $r = 4000$ kms. $= 4 \times 10^8$ cms., the convective transport will predominate if v_r greatly exceeds 5×10^{-11} cm./sec. This velocity corresponds to a displacement of only some 16 meters per million years and only 32 kms. in 2 billion years—the lifetime of the earth.

A solution by Pekeris (1935) for the thermal convection currents in an earth of high kinematic viscosity (3×10^{21}c.g.s.) yields convection currents of velocity 3×10^{-9} to 3×10^{-7} cm./sec.—namely, 60 to 6000 fold the limiting value above. Whether convection exists now in the mantle of the earth, or has ever been effective since an early stage when the crust solidified, are still open questions. If convection has existed, its influence upon heat transport very probably has far exceeded that of conduction. But the fact that the heat transport occasioned by vanishingly small convection velocities may far exceed that due to the conductivity process does not, of course, imply that such convective transfers are large. Both types may be, and in fact probably are, exceedingly small.

The problem of a viscous earth cooling primarily by convection is obviously of exceedingly great complexity. The unknown mechanical properties of the magma, the unknown driving forces that may have operated, and the analytical difficulties of the problem are all obstacles of the first magnitude. With the remark, therefore, that convective cooling might easily predominate, we shall leave this subject and consider the cooling problem under the alternative hypothesis that the mantle of the earth has remained essentially rigid.

PART II. THEORY OF THE COOLING OF A RADIOACTIVE EARTH

EQUIVALENT NONRADIOACTIVE EARTH

In the following simple procedure for solving the problem of the cooling [7] of a radioactive earth, the nonhomogeneous partial differential equation is reduced to one of homogeneous type by considering the radioactive earth as thermally equivalent to a nonradioactive one with suitable altered initial temperature.[8] In essence, the method consists merely in adopting the final steady state temperature as a new zero of reference

[7] For brevity, the word "cooling," in the present paper, will be used to denote temperature change of either sign—*i.e.*, it may mean either "cooling" or "heating."

[8] The author has recently noted that essentially this device was used by Jeffreys (1921, p. 122-149; 1916, p. 575-591) in connection with the cooling of a flat earth at shallow depth.

and solving for the departure from this temperature. In the notation of equation (1) the heat-flow equation for a symmetrical solid earth in which the heat generation is $H(r)$ cal./cc.sec. is

$$\rho c \frac{\partial T}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(\sigma r^2 \frac{\partial T}{\partial r} \right) + H(r) \quad \ldots\ldots (4)$$

Here, H, ρ, c, and σ may all be functions of r. The boundary conditions are as follows:

(a) The surface temperature, at $r = a$ is uniformly zero. $T = 0$, for $r = a$ ..... (4a)

(b) At the center of the earth, $r = 0$, the gradient $\frac{\partial T}{\partial r}$ vanishes.

$\frac{\partial T}{\partial r} = 0$ for $r = 0$. ..... (4b)

(c) The initial temperature distribution, $T_0(r)$, is assigned.

$$\lim_{t \to 0} T(r,t) = T_0(r) \quad \ldots\ldots (4c)$$

When the radioactive heat term H is essentially constant in time, as it is in the earth, the ultimate steady-state temperature, T_∞, satisfies

$$0 = \frac{1}{r^2} \frac{\partial}{\partial r} \left(\sigma r^2 \frac{\partial T_\infty}{\partial r} \right) + H(r) \quad \ldots\ldots (5)$$

an equation which is readily solved for T_∞ by integration, namely,

$$T_\infty = \int_r^a \frac{1}{\xi \sigma(\xi)} \int_0^\xi H(\eta) \eta^2 d\eta d\xi \quad \ldots\ldots (6)$$

Clearly, T_∞ must satisfy the same boundary conditions at $r = 0$ and $r = a$ as does $T(r,t)$. Denote by $\tau(r,t)$ the difference, $T(r,t) - T_\infty(r)$, –

$$\tau(r,t) \equiv T(r,t) - T_\infty(r) \quad \ldots\ldots (7)$$

and form the difference between Equations (4) and (5). It results that $\tau(r,t)$ satisfies the simple heat-flow equation with the radioactive term absent, namely,

$$\rho c \frac{\partial \tau}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(\sigma r^2 \frac{\partial \tau}{\partial r} \right) \quad \ldots\ldots (8)$$

At the earth's surface $\tau(r,t)$ also vanishes, as does $\frac{\partial \tau}{\partial r}$ at $r = 0$; and at the initial time, $t = 0$, $\tau(0,r) = T_0 - T_\infty$. Thus, $\tau(r,t)$ is precisely the temperature distribution in a nonradioactive earth, cooling to zero from an initial temperature which is the difference between the actual initial temperature and the equilibrium temperature which ultimately would be attained. The cooling problem for the radioactive earth is therefore essentially equivalent to the simple problem of the cooling of a nonradioactive sphere from prescribed initial temperature conditions. No distinction need be drawn, for the present, between that part of the

equivalent initial temperature which is the true initial temperature and that part which, in effect, expresses the presence of radioactive heat. It is worthy of remark that the initial equivalent temperature $\tau(r,0)$ involves all three of the major uncertainties in the problem of the cooling earth—namely, this initial condition is the difference between the true initial temperature distribution, T_0, at the time of solidification, and the ultimate distribution, T_∞, which in turn is determined only if both the heat generation and the conductivity be known at all depths. Familiar results in the classical heat-flow problem of the sphere may now be applied in the examination of the present problem. For convenience the transient temperature, $\tau(r,t)$, for the equivalent problem will be referred to as the "equivalent" temperature.

SOLUTION FOR THE EQUIVALENT TEMPERATURE

General statement.—Separation of the variables in Equation (8) by the standard substitution

$$\tau(r,t) = \sum_{n=1}^{\infty} A_n R_n(r) T_n(t) \quad \ldots\ldots (9)$$

leads to the following boundary value problem for $R_n(r)$

$$\frac{\partial}{\partial r}\left(r^2\sigma \frac{\partial R_n}{\partial r}\right) = -\beta^2{}_n r^2 \rho c R_n \quad \ldots\ldots (10a)$$

$$R_n(a) = 0 \quad \ldots\ldots (10b)$$

$$\frac{\partial R_n}{\partial r} = 0 \text{ at } r = 0 \quad \ldots\ldots (10c)$$

It is well known (*see* for example Lovitt, 1924, Chap. VI) that the characteristic numbers, $\beta^2{}_n$, for this type of boundary problem are positive and that the fundamental functions of the boundary problem, $R_n(\beta_n,r)$, are of oscillatory type and form a complete normalized orthogonal system.

Similarly, the functions $T_n(t)$ satisfy

$$\frac{\partial T_n}{\partial t} = -\beta^2{}_n T_n \quad \ldots\ldots (11)$$

namely,

$$T_n(t) = exp\,(-\beta^2{}_n t) \quad \ldots\ldots (12)$$

Thus, the initial temperature, at $t = 0$ is, by Equation (9)

$$T(r,0) = \sum_{1}^{\infty} A_n R_n(\beta_n, r) \quad \ldots\ldots (13)$$

where the constants A_n are to be determined by the relation

$$A_n = \int_0^a x^2 \rho(x) c(x)\, \tau(x,o) R_n(\beta_n, x) dx \quad \ldots\ldots (14)$$

The above will suffice as a general outline of the mathematical structure of the cooling problem for a nonhomogeneous radioactive earth.

Amount of cooling, conductivity constant.—The major features of the problem will be retained, and the essence of the matter much more simply arrived at, if it be assumed that the conductivity, σ, and the product ρc in Equation (10a) are constants. In view of the uncertainties about

TABLE 4.—*Cooling after 2 × 10⁹ years*

In an earth of thermal diffusivity .01, for various fundamental modes, $n = 1$ to 20.

n	1	2	3	4	5	6	7
$e^{-.0153n^2}$	.985	.940	.872	.783	.682	.577	.472
n	8	9	10	12	15	20	22
$e^{-.0153n^2}$	.376	.289	.216	.110	.031	.002	.001

the physical properties at depth, this assumption is apparently as realistic as any. With σ and ρc constants, the problem possesses Fourier's (1822) well-known solution,

$$\tau(r\ t) = \sum_1^\infty A_n\ exp\left(-\frac{n^2\pi^2}{a^2}\ kt\right)\frac{sin\ n\pi r/a}{r} \quad \ldots\ldots\ldots\ldots\ldots\ldots (15)$$

where

$$A_n = \frac{2}{a}\int_o^a r\tau(r,\ o)\ sin\ \frac{n\pi r}{a}\ dr \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (16)$$

Equation (15) states that the equivalent initial temperature $\tau(r,t)$ is expressed as the sum of fundamental modes, of type $A_n\ \frac{sin\ n\pi r/a}{r}$, each of which cools (or *heats*, if the sign of A_n be negative) to the ultimate temperature zero in accordance with its individual exponential, $exp\left(-\frac{n^2\pi^2}{a^2}\ kt\right)$. Several of the fundamental modes—namely those for $n = 1$, 2, and 10—are plotted in Figure 1, each with amplitude $A_n = \frac{a}{n\pi}$. In the case of the earth, the "wave length" of the temperature perturbation corresponding to $n = 10$ is $\frac{2 \times 6{,}370}{10} = 1274$ kms. The amount of cooling after 2 billion years in a solid earth of diffusivity 0.01 which is associated with each of the modes $n = 1$ to 10, inclusive, and $n = 12$, 15, and 20 is listed in Table 4. The cooling is here expressed as the ratio of the present amplitude of the given mode to its original amplitude.

It is evident that relatively little cooling has occurred throughout the whole lifetime of the earth in the case of the longer perturbations ($n = 5$ or less). All perturbations of wave length greater than 1800 kms. ($n < 7$) retain more than half their original amplitude, and the fundamental mode, $n = 1$, has lost only 1½ per cent of its original temperature.

But when $n > 10$, corresponding to perturbations of wave length 1250 kms. or less, the present temperature amplitudes in the assumed model are less than 20 per cent of their original values; and original tempera-

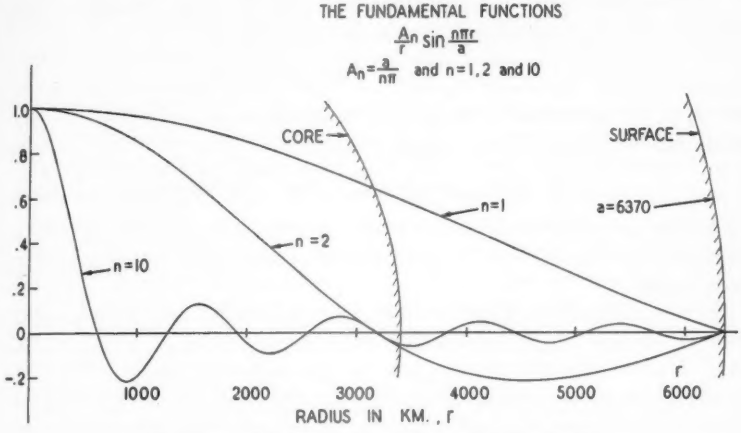

FIGURE 1.—*Fundamental functions for* n = 1, 2, 10

ture perturbations of wave length 800 kms. ($n = 15$) or less have now almost entirely disappeared. The above statements, of course, depend upon the value, 0.01, assumed for the diffusivity. However, since the diffusivity enters the exponent only as a first power, whereas the factor n is squared, the amount of cooling is decidedly more sensitive to wave length than to the diffusivity.

MODES OF MAXIMUM PRESENT RATE OF COOLING

It has been noted that temperature perturbations of short wave length now retain only a small part of their original temperature and have in effect cooled completely, whereas those of long wave length have cooled scarcely at all. Evidently, there exist modes of an intermediate wave length for which the cooling rate is now a maximum. The modes of maximum present cooling rate are, indeed, simply found. Let $\tau_n(r,t)$ represent the *nth* fundamental mode,

$$\tau_n(r, t) = A_n \frac{\sin n\pi r/a}{r} \exp\left(-\frac{n^2\pi^2}{a^2} kt\right) \quad \ldots\ldots (17)$$

The rate of cooling, $\frac{\partial \tau}{\partial t}$, of this mode is

$$\frac{\partial \tau}{\partial t} = A_n \frac{\sin n\pi r/a}{r} \left(\frac{s\, e^{st}}{t}\right) \quad \ldots\ldots (18)$$

where

$$s = -\frac{n^2\pi^2}{a^2} kt. \quad \ldots\ldots (19)$$

The rate, $\frac{\partial \tau}{\partial t}$, is a maximum with respect to s when $s = 1.0$, and, if $t = 6.3 \times 10^{16}$ seconds (2 billion years) and $k = 0.01$, this implies that n is 8 or 9—that is, the mode which would now be cooling most rapidly in a solid uniform earth has a half wave length of about 750 km. The present rate of change of temperature in this mode is

$$\frac{\partial \tau}{\partial t} = \frac{.368}{t} \tau_o = 5.84 \times 10^{-18} \tau_o \ldots\ldots\ldots\ldots\ldots\ldots (20)$$

where τ_0 represents the initial state of the mode, $\tau_o = A_n \frac{\sin n\pi r/a}{r}$. The temperature change per 100 million years is

$$\Delta t \frac{\partial \tau}{\partial t} = 5.84 \times 10^{-18} \times 3.15 \times 10^{16} \tau_o = 1.8 \times 10^{-2} \tau_o \ldots\ldots\ldots (21)$$

If τ_0 had an initial amplitude of 1000°, the present rate of temperature change would be about 18° C. per hundred million years, which is clearly very small.

In a similar way it may be readily shown that the maximum present rate of change of the surface thermal gradients occurs for the modes $n = 5$ or 6. (It is here assumed, for purposes of comparing rates of change, that all the amplitude factors, A_n, are equal; it is also assumed that $k = 0.01$ and $t = 6.3 \times 10^{16}$, as before). The maximum rate turns out to be small. After 100 million years, the present gradient for the mode of maximum rate of change, $n = 5$ or 6, will have changed by only about 2 per cent.

LARGE THERMAL INERTIA OF EARTH, EXAMPLES

General considerations.—The fact that the fundamental modes for small values of n have suffered so small a percentage change in temperature during the earth's lifetime is, of course, a direct result of the earth's large size and small thermal diffusivity. Indeed, in 1822 in his original solution of the problem of the cooling sphere, Fourier noted that, "the final cooling is very slow in spheres of great diameter." However, the magnitude of the heat capacity of the earth and its importance to the present problem are not always fully appreciated by geologists. Accordingly, it seems well to emphasize this feature by mention of several elementary examples which will convey in a simple, vivid way a concept of the magniture and effects of the earth's thermal inertia.

Heat capacity of earth.—If the earth's mean density is 5.5 and specific heat is about .3, then the earth's thermal capacity is 1.8×10^{27} cal./deg. The observed rate of heat loss is about 1.2×10^{-6} cal./cm.^{2} sec. or over the entire surface, about 6.1×10^{12} cal./sec. This loss rate, maintained throughout the lifetime of 2 billion years, would suffice to cool the entire earth by only some 200° C.

To make available the large supply of original heat, it is only necessary that adequate means, in the form of suitable initial thermal gradients and values of the conductivity at depth, be provided for transporting heat to the surface. It is certain that a nonradioactive earth model could be designed, if desired, which would be compatible with all the thermal conditions on the problem, as we now know them—namely, the conditions (a) of zero surface temperature (b) of agreement with the known surface heat-flux and gradients at the present elapsed age of the earth, and (c) the looser, more vague condition of "reasonableness" for the initial internal temperature distribution, and for the values of the thermal conductivity and diffusivity at depth. However, in view of the known existence of large amounts of radioactive heat generation, a proposed model for a nonradioactive earth possesses little interest. The point which it is desired to emphasize is merely that the heat capacity of the earth is *large*—great enough to supply the total estimated heat losses without aid from radioactivity and without notable loss in mean temperature.

Slowness of thermal diffusion in earth.—On a uniform solid earth having the thermal diffusivity observed in surface rocks, let the surface temperature be suddenly changed, and maintained thereafter at a new value. In such a case it is readily found that the temperature of the earth's center will remain essentially unaffected (that is to say, to an extent less than 10 per cent of the imposed surface temperature change) for 200 billion years, or about 100-fold the earth's lifetime. If the diffusivity at depth be 10-fold the surface value, the diffusion process is correspondingly speeded; an interval of 10 times the earth's lifetime must elapse before the 10 per cent change is produced. The size of the earth is so great, and the thermal diffusivity of its materials apparently so small, that its interior is nearly thermally independent of its exterior.

Cooling rates by inspection of heat-flow equation.—A curve purporting to represent present temperature distribution in the solid earth must satisfy the following four obvious conditions. At the surface it has (1) the value zero, with (2) an initial gradient to correspond with observed values, (3) at the center, the slope is zero, and (4) the central temperature is unknown, but "reasonable." By "reasonable" is meant a temperature measured in thousands of degrees, say between 3000° C. and 15,000° C. If an additional assumption (which seems universally to have been made) be accepted—namely, that the temperature always increases with depth, but at a diminishing rate—then the smooth curves which represent any temperature distribution of the character described must necessarily have small values of the curvature. In curves which

have been proposed by Jeffreys, Adams, and Gutenberg, and in arbitrary curves sketched by the writer (drawn for a central temperature of 5000° C.), the values of the term $\frac{\partial^2 T}{\partial r^2}+\frac{2}{r}\frac{\partial T}{\partial r}$ at depths of 500 km. or greater are about -1 to -4×10^{-13}. Thus, in accordance with the heat-flow equation (Equation 8), cooling by conduction occurs at the approximate rate $\frac{\partial T}{\partial t}=-k$ (1 to 4×10^{-13}). This, with $k = .01$, corresponds to a temperature drop of between 30° C. and 120° C. per billion years. At greater depths the cooling so estimated is decidedly less. Such small rates of heat loss by conduction would be compensated by a radioactive heat generation of 1 to 4×10^{-15} cal./cc.sec.—namely, about 1 per cent of that observed in surface basalts or granites. Thus, if such an earth is cooling at all at depth, the rate must be exceedingly slow. On the contrary, it is clearly possible for *heating* to occur at a rate more than tenfold that mentioned. This would mean temperature increases of 300° C. to 1200° C. or more per billion years.

MANTLE PROBABLY SOLIDIFIED FROM BOTTOM UP

The above three elementary examples indicate that cooling rates by conduction, in the deep interior, must necessarily have always been very small. This conclusion is, of course, a matter of common knowledge and is in agreement with those which have already been noted in connection with the cooling of the fundamental modes. Such results show that the solid mantle of the earth at depth cannot have cooled significantly during the earth's lifetime. Therefore, if the mantle was initially molten, it probably solidified progressively from the bottom up, beginning at the boundary of the molten core. The initial solidification of a great thickness of the mantle either at the surface or at some intermediate depth would have introduced effective thermal insulation. Subsequent cooling of the magma below would have been essentially halted thereby; and unless by chance this magma had already nearly cooled to its freezing point at the time when solidification began above, it would have remained fluid until today. The hypothesis that the mantle was once fluid, and is now almost everywhere solid, therefore implies either (1) that freezing began at the bottom, with subsequent rapid cooling of the magma above by convection, or (2) that the mantle had cooled nearly to its freezing point at all depths below that where solidification first occurred and was thereby able to congeal without much additional loss of heat. Clearly hypothesis (1) implies a less delicately balanced type of condition, and a priori possesses greater probability of truth. Furthermore the rapid cooling implied by hypothesis (1) would not be materially affected by the presence of radioactivity, whereas hypothesis

(2) is tenable only if the heat generation at depth be less than a few per cent of that in surface rocks, for otherwise no cooling could occur.

EFFECT OF FLUIDITY OF THE CORE

The preceding discussions neglected consideration of the probable fluidity of the core. Fluidity, aided possibly by superior thermal conductivity, would tend to preserve uniformity of the core's temperature. It would, in effect, make more available the large thermal capacity of the core for maintaining constancy of temperature at the inner boundary of the mantle. However, the effects of fluidity in the core probably are insignificant because the temperatures deep in the mantle in any case change only slowly with time for the modes of long wave length. In the fundamental mode it has been seen that the cooling for a solid earth amounts to only 1 or 2 per cent.

EQUILIBRIA TEMPERATURES AND ASSOCIATED HEAT GENERATION

General statement.—The answer to the important question—Is the earth heating or cooling at depth?—clearly depends upon knowledge of two things: (1) the initial temperature distribution in the earth, and (2) the equilibrium temperature ultimately to be established by the radioactive heat. If the former temperature exceeds the latter everywhere, the earth is cooling everywhere; if the latter exceeds the former, it is heating everywhere; and if the curves representing the two distributions intersect, cooling occurs at some depths and heating at others. The rate of heating or cooling depends upon the difference between the two curves. If by chance the initial state nearly coincided with the final one, the internal temperature would, of course, remain essentially unchanging with time.

According to the process for the formation of the mantle outlined in Part I, the initial temperatures in the solid mantle are those of the depth freezing-point curve of its materials in the physical and chemical environment under which the freezing occurred. This curve is certainly not satisfactorily known, but the initial temperatures are generally assumed to increase from about 800° at the base of the crust to several thousand degrees at the base of the mantle.

The final temperatures depend in a simple way (*see* Equation 6) upon the radioactivity and conductivity and may be computed for a range of different hypotheses concerning these quantities. Examples of steady-state temperatures for some dozen earth models of different types will be discussed. These models are of the following general classes:

(1) Layered Earths, in which the radioactivity and the conductivity is uniform in shells but changes discontinuously at the shell boundaries.

(2) Earths of constant conductivity, in which the radioactivity decreases exponentially with depth.

(3) Earths of constant conductivity in which the radioactivity varies in accordance with certain other useful formulae.

Layered earths.—The surface heat flow in the layered earths chosen for computation has generally been set approximately equal to that observed today in the crust. This clearly is an arbitrary choice—the flux today may still be notably below its equilibrium value, and the temperatures computed in the examples may accordingly be too small at depth. The two outermost layers in the earth models are intended to represent the crust. The deeper layers bear no relation to actual earth structure. They represent merely assigned transitions in the heat generation and thermal conductivity for the purpose of approximating a desired type of variation for these quantities. The radioactivity is assumed to decrease rapidly in successive layers at depth, and the conductivity is chosen to increase. Since the temperature increments depend merely upon the ratios Hi/σ_i (*see* Equation 22), the type of layering chosen tends to minimize the steady-state interior temperatures.

Let r_i denote the outer radius of the shell throughout which the conductivity is σ_i, and the heat term H_i. When H_i and σ_i are constants, integration of Equation (5) yields, for the temperature $T_i(r)$ in the ith layer,

$$C_{2i} + C_{1i}\, r = r\, T_i(r) + \frac{H_i r^2}{\sigma_i 6} \qquad (22)$$

The constants C_{1i}, C_{2i} are determined by the boundary conditions at the interfaces of the shells, which are

$$T_{i+1} = T_i \quad at\ r = r_i \qquad (22a)$$

$$\sigma_{i+1} \frac{\partial T_{i+1}}{\partial r} = \sigma_i \frac{\partial T_i}{\partial r} \quad at\ r = r_i \qquad (22b)$$

The constants C_{1i} and C_{2i} are conveniently evaluated successively from the surface down, starting with temperature zero, and an assigned heat flux at the outer surface, $r = a$. The total heat generation in all layers is clearly fixed by the assigned surface flow at $r = a$.

In Table 5 are listed the steady-state temperatures produced at the depths indicated, in earths of the specified layer thicknesses, heat generation, and thermal conductivity. The influence of the thermal conductivity is separately indicated by including comparison temperatures for the case of constant conductivity, $\sigma = 0.01$. The majority of the functions tabulated in Table 5 are shown plotted in Figure 2. In comparing these plots, it should be noted that the temperature and heat generation scales in cases (1) and (2) differ from the rest; the scales in the cases (3) to (11), inclusive, are the same.

TABLE 5.—*Examples of equilibria temperature distributions*

For various types of radioactive earths

CASE (1) SURFACE HEAT FLUX, -1.375×10^{-6} CAL./CM.^{2} SEC.

Depth km.	Heat cal./cc. yr.	Heat cal./cc. sec.	Thermal conduct.	Depth km.	Temp. °C.	Temp. $\sigma = .01$
0–10	25.5	8.1	.0055	0	0	0
10–32	8.1 $\times 10^{-6}$	2.6 $\times 10^{-13}$	.0062	10	177	97
32–6370	0	0	.....	32	279	160
				Center	279	160

CASE (2) EXPONENTIAL DECREASE IN RADIOACTIVITY WITH DEPTH SURFACE HEAT FLUX, -1.2×10^{-6} CAL./CM.^{2} SEC. CONDUCTIVITY $-.006$

Depth km.	Heat cal./cc. yr.	Heat cal./cc. sec.	Temp. °C.
0	14.5	4.6	0
6.37	11.3	3.6	117
12.74	8.9	2.8	205
25.4	5.5	1.7	330
63.7	1.3 $\times 10^{-6}$	.4 $\times 10^{-13}$	484
127.4	.116	.037	528
254	.000	0	532
Center	0	0	532

CASES (3), (4) AND (9) SURFACE HEAT FLUX AS IN (1)

Depth km.	Heat cal./cc. yr.	Heat cal./cc. sec.	Conductivity (3)	(4)	(9)	Temperature (3)	(4)	(9)	Temp. $\sigma = .01$	Depth km.
0–15	12.1	3.8	.0055	.0055	.0055	0	0	0	0	0
15–40	4.0	1.1	.0124	.0062	.0062	297	297	297	163	15
40–200	.178 $\times 10^{-6}$	.057 $\times 10^{-13}$	.024	.024	.008	428	559	559	326	40
200–570	.080	.025	.030	.03	.01	734	865	1477	1060	200
570–6370	.061	.020	.15	.15	.05	1231	1362	2969	2552	570
						1954	2085	5137	13,392	Center

CASE (5) OCEANIC CRUST. GRANITIC LAYER ABSENT. OCEAN—4.3 KM. DEEP (COMPARE CASE (6)). SURFACE HEAT FLUX, -1.1×10^{-6} CAL./CM.^{2} SEC.

Depth km.	Heat cal./cc. yr.	Heat cal./cc. sec.	Thermal conduct.	Temp. °C.	Temp. $\sigma = .01$	Depth km.
4.3– 34.3	2.7	.86	.0062	0	0	4.3
34.3–194	1.31	.42	.008	475	294	34.3
194 –564	.16 $\times 10^{-6}$	.050 $\times 10^{-13}$	.010	1556	1158	194
564 –6366	.006	.002	.05	2057	1659	564
				2271	2729	Center

CASES (6) AND (7) SURFACE HEAT FLUX, -1.375×10^{-6} CAL./CM.^{2} SEC. AS IN (1)

Depth km.	Heat cal./cc. yr.	Heat cal./cc. sec.	Conductivity (6)	(7)	Temperature (6)	(7)	Temp. $\sigma = .01$	Depth km.
0–10	8.64	2.7	.0055	.0055	0	0	0	0
10–40	2.7	.86	.0062	.0062	225	225	124	10
40–200	1.31 $\times 10^{-6}$	.42 $\times 10^{-13}$	.08	.08	700	700	418	40
200–570	.16	.051	.01	.0048	1781	1781	1282	200
570–6370	.006	.002	.05	.05	2282	2782	1783	570
					2496	2996	2853	Center

TABLE 5.—*Examples of equilibria temperature distributions*—Continued

CASE (8) OCEANIC CRUST. GRANITIC LAYER ABSENT. OCEAN 4.3 KM. DEEP. (COMPARE CASE (9)). SURFACE HEAT FLUX, $-.805 \times 10^{-6}$ CAL./CM.^{2} SEC.

Depth km.	Heat cal./cc. yr.	Heat cal./cc. sec.	Thermal conduct.	Temp. °C.	Temp. $\sigma = .01$	Depth km.
4.3–29.3	4.0	1.1	.0062	0	0	4.3
29.3–189.3	.178 $\times 10^{-6}$	.057 $\times 10^{-12}$	.008	262	162	29.3
189.3–559.3	.08	.025	.010	1180	896	189.3
559.3–Center	.061	.020	.050	2672	2388	559.3
				4840	13,228	Center

CASE (10) SURFACE HEAT FLUX, -2.75×10^{-6} CAL./CM.^{2} SEC. (COMPARE CASE (1)).

Depth km.	Heat cal./cc. yr.	Heat cal./cc. sec.	Thermal conduct.	Temp. °C.	Temp. $\sigma = .01$	Depth km.
0–10	25.5	8.1	.0055	0	0	0
10–40	8.1 $\times 10^{-6}$	2.6 $\times 10^{-12}$	.0062	427	235	10
40–200	2.4	.76	.0080	1187	706	40
200–Center	0	0	.....	2229	1540	120
				2387	1666	200
				2387	1666	Center

CASE (11) UNIFORM RADIOACTIVITY, $-.179 \times 10^{-6}$ CAL./CC. YR. $= .057 \times 10^{-12}$ CAL./CC. SEC.
UNIFORM CONDUCTIVITY, $-.006$
SURFACE HEAT FLUX, -1.2×10^{-6} (AS IN (2)).
TEMPERATURE $= 63{,}700 \left(1 - \frac{r^2}{a^2}\right)$

Depth km.	Temp. °C.	Temp.	r/a
0	0	12,100	.9
15.9	318	32,500	.7
31.8	637	47,800	.5
47.8	955	61,200	.2
64.3	1274	63,700	.0
128.5	2550		
194.0	3820		
327.0	6370		

The cases (1) to (11) are arranged in order of increasing radioactivity at depth, and so involve progressively higher final internal temperatures. Except as will be noted, the surface heat flux is always either 1.2×10^{-6} or 1.375×10^{-6} cal./cm^{2}sec. Cases (5) and (8) are the same as cases (6) and (9), respectively, except that the "granitic" layer has been omitted, to obtain an earth of "oceanic" type. The surface heat flow is correspondingly reduced from 1.375×10^{-6} to 1.1×10^{-6} cal./cm.^{2}sec. in case (5) and to 0.80×10^{-6} in case (8). In case (1), no heat generation is assumed below a depth of 32 kms. Case (10) is like case (1) except for the addition of heat generation at depth, in particular in the zone 40 km. to 200 km. where it is assumed that the heat generation is 0.76×10^{-13} cal./cm.^{2}sec., or midway between that of the Deccan-

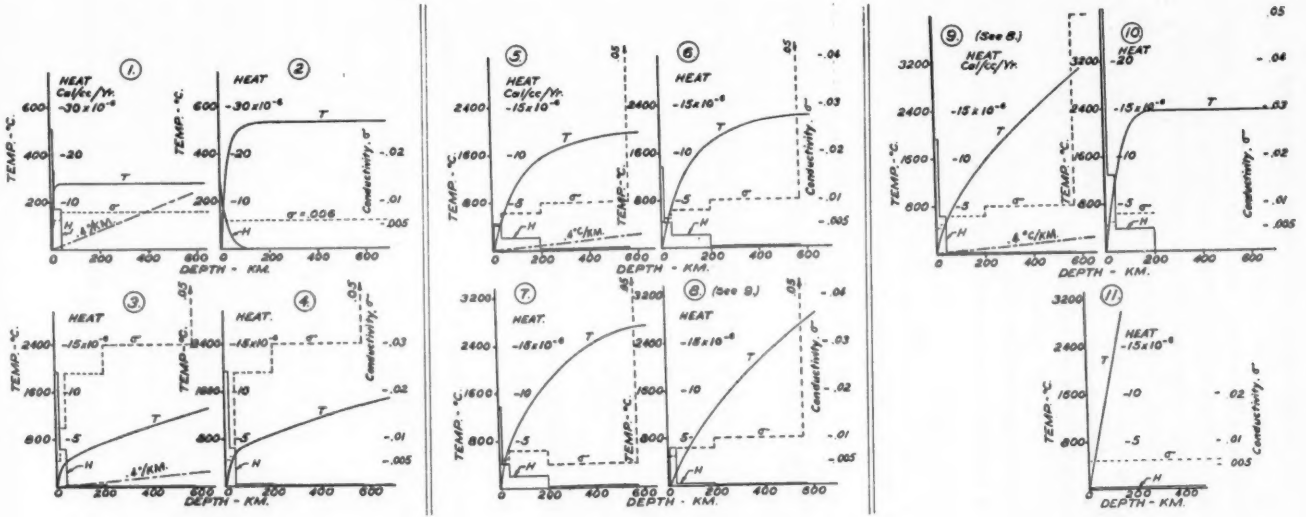

FIGURE 2.—*Examples of equilibria temperature distributions for various types of radioactive earths*

Columbia River basalt average, and that for stony meteorites. The equilibrium surface heat flux is thereby raised to 2.75×10^{-6} cal./cm.^{2} sec., and the central temperatures from 280° C. to 2390° C. A further illustration of the high ultimate temperatures which may be produced by a minor content of radioactive heat is seen in case (11). A uniform radioactivity of about 6 per cent that observed in the surface rocks lowest in radioactivity leads to ultimate central temperatures of over 60,000° C.

The exponential distribution of radioactivity, of which case (2) is an example, is considered in greater detail in the following paragraph.

Exponential distributions.—The exponentially decreasing type of heat distribution is often assumed (Ingersoll and Zobel, 1913; Lowan, 1933; Van Ostrand, 1940). If the heat generated is

$$H = H_o \exp\left[-\beta(a - r)\right] \quad \ldots\ldots(23)$$

then, by Equation (6), the steady-state temperature in an earth of conductivity σ is:

$$T = \frac{H_o}{\sigma\beta^2}\left[1 - \frac{2}{\beta a} - \left(1 - \frac{2}{\beta a y}\right)\exp\left\{-\beta a\,(1 - y)\right\} + \frac{2}{\beta a}\exp\left(-\beta a\right)\right] \quad \ldots\ldots(24)$$

where $y \equiv r/a$.

At the center,

$$T_o = \frac{H_o}{\sigma\beta^2}\left[1 - \frac{2}{\beta a} + \left(1 + \frac{2}{\beta a}\right)\exp\left(-\beta a\right)\right] \quad \ldots\ldots(25)$$

The equilibrium heat flux at the surface is

$$-\sigma\frac{\partial T}{\partial r} = \frac{H_o}{\beta}\left\{1 - \frac{2}{\beta a}\left[\frac{\beta a - 1 + \exp\left(-\beta a\right)}{\beta a}\right]\right\} \quad \ldots\ldots(26)$$

Usually the arbitrary constant, β, is chosen large enough to render negligible the exponential term $\exp(-\beta a)$.

In case (2) of Figure 2a the plotted temperature corresponds to the values

$$H_o = 4.6 \times 10^{-13} \text{ cal./cc. sec.}$$
$$\sigma = .006$$
$$\beta a = 242, \text{ i.e., } \beta^2 = 1.45 \times 10^{-13}$$

The corresponding surface heat flow is 1.2×10^{-6} cal./cm.^{2}sec.—*i. e.*, about that observed at present, but the radioactivity in the example decreases exceedingly rapidly from a surface value of 4.6×10^{-13}, corresponding approximately to the Evans-Goodman value in granite, to less than 10 per cent of this value at a depth of only 125 km. The ultimate temperature is accordingly nearly constant at depths below 100 km., at the low value 530° C. Evidently such an earth would be cooling

at depth. With other, and possibly more tenable, values for the constants, a high ultimate internal temperature is readily obtained. For example, let $H_0 = 10^{-13}$, corresponding to the heat generation in Deccan traps and Columbia River basalts; let $\sigma = 0.01$. $\beta a = 19$. In this case the ultimate central temperature of 10,000° is 99 per cent attained at a depth of 1400 km. The equilibrium heat flux is 3×10^{-6}, and the radioactivity drops to 10 per cent of its surface value in a depth of 700 km. Because of the high ultimate internal temperature, this example probably represents a heating earth.

The transient temperatures for the exponential type of distribution have been expressed and computed by Lowan (1933) and Van Ostrand (1940). Lowan's expression for the temperature rise due to the generation of radioactive heat is

$$T(r, t) = \frac{2}{\pi} \frac{H_0}{\sigma} \frac{a^2}{r} \sum_1^\infty (-)^{n+1} \frac{\sin n\pi r/a \left[1 - exp\ (-\frac{n^2\pi^2}{a^2} kt)\right]}{n\ (\beta^2 a^2 + n^2\pi^2)} \quad \ldots\ldots (27)$$

Lowan computed the transient temperatures from Equation (27) for the values $H_0 = 10^{-12}$ cal./cc.sec., $k = 0.005$ and 0.007, $\sigma = 0.006$, $\beta a = 448$, $\beta = 6.66 \times 10^{-7}$, and $t = 1.6 \times 10^9$ years. These values correspond to an ultimate surface heat flow of 1.5×10^{-6}, and a steady central temperature of 375°. The surface gradient in this example has essentially achieved its ultimate value, but the temperature at a depth of 200 km. is still only about 50 per cent of its final value, whereas at 30 km. depth it is about 1⅓ times the steady value.

The sum in formula (27) converges very slowly, especially for small values of the depth, $a - r$, and for large values of βa; in one of Van Ostrand's examples ($\beta a = 6{,}370$) the series was summed to 6740 terms, for the case $a - r = 1$ km., to 2540 terms for the depth 50 km., and to 160 terms for depth 200 km. The difficulty arises partly [9] from the fact that the initial coefficients decrease, when βa is large, only as $1/n$ until πn approaches βa. The assumption of an exponential distribution is not an especially convenient one, and other, more tractable choices are recommended.

Certain convenient types of heat distributions.—The following four distributions of radioactivity lead to simply computable expressions both for the steady-state temperatures and for the transient temperatures. Moreover, when taken in combination, with amplitude factors of appro-

[9] For an exponential distribution in which the heat generation is concentrated near the surface, it is well known that the solution at shallow depths is conveniently computed by adopting the approximation of a flat earth.

priate size and sign, they may be used to approximate a wide variety of suitable heat-generation functions. These functions are the following:

(1) The fundamental functions,

$$H = H_o \frac{\sin n\pi r/a}{n\pi r/a} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (28)$$

to which correspond the ultimate temperatures

$$T = T_o \frac{\sin n\pi r/a}{n\pi r/a} \ , \ T_o = \frac{a^2}{n^2\pi^2} \frac{H_o}{\sigma} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (29)$$

Here H_0 denotes the rate of heat generation at the center of the earth, and T_0 its ultimate central temperature.

(2) The distribution

$$H = \frac{H_o}{3}\left(\cos\frac{n\pi r}{2a} + \frac{2\sin\dfrac{n\pi r}{2a}}{\dfrac{n\pi}{2} r/a} \right) \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (30)$$

to which corresponds

$$T_\infty = T_o \cos\frac{n\pi r}{2a} \ , \ T_o = \frac{4}{3}\frac{a^2}{n^2\pi^2}\frac{H_o}{\sigma} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (31)$$

The mode for $n = 1$ is especially useful, and in subsequent reference to cases (1) and (2) the value $n = 1$ will be presumed, unless otherwise indicated.

(3) The constant distribution,

$$H = H_o \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (32)$$

to which corresponds

$$T = T_o\,(1 - r^2/a^2) \ , \ T_o = \frac{a^2}{6} H_o/\sigma \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (33)$$

(4) The heat distribution which is constant throughout a central core of radius r_1 and zero elsewhere. Thus,

$$H = H_o, 0 < r < r_1; H = 0, r > r_1 \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (34)$$

To this corresponds the steady-state temperature

$$T_{1\infty} = \frac{H_o r_1^2}{\sigma}\frac{1}{6}\left\{ 1 + 2\left(1 - \frac{r_1}{a}\right) - \left(\frac{r}{r_1}\right)^2 \right\}, \ 0 \ll r \ll r_1 \quad \ldots\ldots\ldots\ldots\ldots (35a)$$

$$T_{2\infty} = \frac{H_o r_1^2}{\sigma}\frac{1}{3}\left\{ \frac{1}{r} - \frac{1}{a} \right\}, \ r_1 \ll r \ll a, \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots (35b)$$

(4a) From cases (3) and (4) is obtained by subtraction the steady temperature distribution due to heat generation uniform throughout a superficial shell of thickness $h = a - r_1$. This is a special case of the steady temperatures in layered radioactive earths which have been listed in Table 5.

In Figure 3, in solid line, are plotted the first three temperature functions above, each with the same central temperature, and the correspond-

ing heat generation functions in dotted line, for comparison. The three temperature curves are nearly alike, and they obviously are not sensitive to the differences in their respective generating functions.

The steady-state temperatures illustrated in Figures 2 and 3 are in

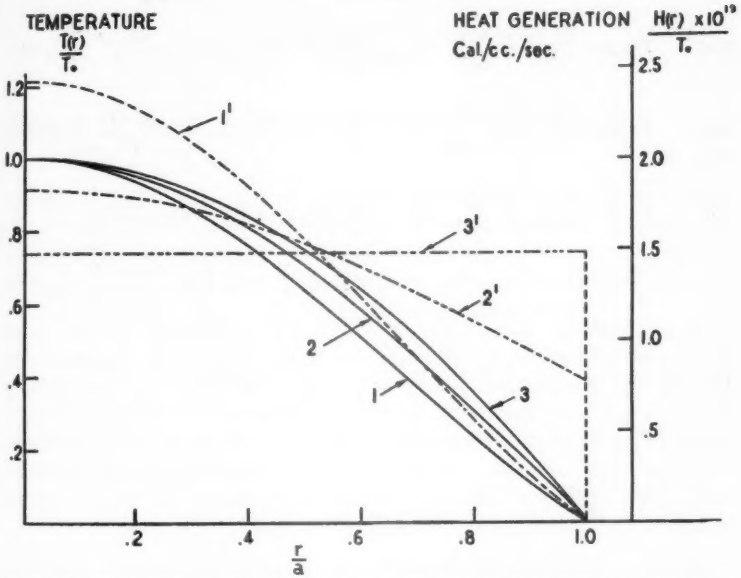

FIGURE 3.—*Certain steady-state temperatures and associated heat-generation functions*
T_0 = temperature at center; H_0 = heat generation at center

many instances very large at depth. If the assigned amounts of radioactivity (which are very small, judged from observational experience) be accepted, then the probability is strong that the earth is heating at depth. It is also apparent from inspection of these temperatures that a curve representing the initial temperature in the mantle might intersect at depth, in appropriate cases, the steady-state temperature curves of Figures 2 and 3, thus leading to cooling at certain depths and heating at others. Whether distributions of radioactivity of the small concentrations suggested in these examples are compatible with the observable thermal data at the surface will be considered hereinafter.

TRANSIENT TEMPERATURES

General remarks.—Formulae for the transient temperatures $T(r,t)$ existing in earths being heated from zero by the above four types of

radioactive distributions—Equations (28), (30), (32), and (34) respectively—are readily calculated.[10] (*See* Equations (15) and (16).) They are as follows:

$$\text{(1a)} \qquad T(r,t) = \frac{H_o}{\sigma}\frac{a^2}{n^2\pi^2}\frac{\sin n\pi r/a}{n\pi r/a}\left[1 - exp\left(-\frac{n^2\pi^2 kt}{a^2}\right)\right] \ldots\ldots\ldots (36)$$

$$\text{(2a)} \qquad T(r,t) = \frac{H_o}{\sigma}\frac{64}{3\pi^4}\frac{a^3}{r}\sum_1^\infty \frac{(-)^{n+1}\,2n\sin n\pi r/a}{[4n^2-1]^2}\left[1 - exp\left(-\frac{n^2\pi^2 kt}{a^2}\right)\right] \ldots (37)$$

$$\text{(3a)} \qquad T(r,t) = \frac{H_o}{\sigma}\frac{2}{\pi^3}\frac{a^3}{r}\sum_1^\infty \frac{(-)^{n+1}\sin n\pi r/a}{n^3}\left[1 - exp\left(-\frac{n^2\pi^2 kt}{a^2}\right)\right] \ldots (38)$$

$$\text{(4a)} \qquad T(r,t) = \frac{H_o}{\sigma}\frac{2}{\pi^4}\frac{a^3}{r}\sum_1^\infty \frac{(-)^{n+1}}{n^4}\left\{\sin n\pi h/a + n\pi(1-h/a)\cos n\pi h/a\right\}$$

$$\sin n\pi r/a\left[1 - exp\left(-\frac{n^2\pi^2 kt}{a^2}\right)\right] \ldots\ldots (39)$$

In the formulae (37), (38), and (39), the convergence of the series is satisfactorily rapid, by virtue of the factor $1/n^3$ in the typical term. Clearly the transient temperatures for earths of constant conductivity in which the radioactivity is distributed uniformly in shells are readily found by forming the appropriated sums and differences of formulae of the type (38) and (39). Similarly, obvious combinations of formulae (36), (37), (38), and (39) may be formed to obtain the transient temperatures for a wide variety of radioactive heat distributions.

Special case. Simple relation between transient temperatures and heat term.—If the heat-generation function vanishes at the surface, then its corresponding transient temperature function may be written down at once, in terms of the coefficients expressing the heat-generation function. To see this, introduce the auxiliary heat function, $h(r)$, defined by

$$\rho ch(r) = H(r) \ldots\ldots (40)$$

Then both $h(r)$ and $T_\infty(r)$ satisfy conditions similar to those (*see* Equations 10a, 10b, 10c) determining the fundamental functions, R_n, of the cooling problem; namely, $h(r)$ and $T_\infty(r)$ satisfy

$$\frac{\partial}{\partial r}\left(r^2\sigma(r)\frac{\partial T_\infty}{\partial r}\right) + r^2\rho(r)c(r)h(r) = 0 \ldots\ldots (41)$$

$$T_\infty(a) = 0\,, \qquad h(a) = 0 \ldots\ldots (41a)$$

$$\frac{\partial T_\infty}{\partial r} = 0 \quad at\ r = 0. \qquad \frac{\partial h}{\partial r} = 0 \quad at\ r = 0 \ldots\ldots (41b)$$

[10] For computing temperatures at shallow depths, z, it is desirable to use the equality, $\sin\frac{n\pi r}{a} = (-)^{n+1}\sin\frac{n\pi z}{a}$, in formulae 37, 38 and 39.

Clearly $h(r)$ (if continuous) and $T_\infty(r)$ may be expanded in terms of the fundamental functions, R_n. Thus,

$$T_\infty = \sum_1^\infty A_n R_n \quad \text{.....(42)}$$

$$h = \sum_1^\infty C_n R_n \quad \text{.....(43)}$$

From inspection of Equations (10c) and (41) it is evident that

$$C_n = \beta^2_n A_n \quad \text{.....(44)}$$

Thus, when the radioactivity vanishes at the surface, the simple relation (44) holds between the amplitudes A_n of the elementary modes of the steady-state temperature distribution and those, C_n, expressing the distribution of radioactive heat. More specifically, if the initial temperature is uniformly zero, then the transient temperature is

$$T(r,t) = \sum_1^\infty \frac{C_n}{\beta^2_n} R_n \left(1 - exp(-\beta^2_n t)\right) \quad \text{.....(45)}$$

In case both σ and ρc are constants, formula (45) reduces to one similar to Equation (36), namely,

$$T(r,t) = \sum_1^\infty \frac{H_n}{\sigma} \frac{a^3}{n^3\pi^3} \frac{sin\ n\pi r/a}{r} \left[1 - exp\left(-\frac{n^2\pi^2 kt}{a^2}\right)\right] \quad \text{.....(46)}$$

where the heat function has the expansion

$$H = \sum_{n=1}^\infty H_n \frac{a}{n\pi} \frac{sin\ n\pi r/a}{r} \quad \text{.....(47)}$$

SURFACE HEAT FLOW

Equations (36), (37), (38), and (39) express transient temperatures in earths being heated from zero by the respective heat-generation functions expressed in Equations (28), (30), (32), and (34). To take into account the effect of the earth's initial temperature, $T_1(r,0)$, the addition of a transient term, $T_1(r,t)$ is needed. This term represents the temperature in a nonradioactive earth cooling to zero from the initial state, $T_1(r,0)$. The surface heat flows, F_i, associated with each of the solutions (36), (37), (38), and (39), amended by inclusion of the term $T_1(r,t)$, are respectively

$$F_1 = -\sigma \frac{\partial T}{\partial r}\bigg|_o = \frac{H_o\, a(-)^{n+1}}{n^2\pi^2} \left[1 - exp\left(-\frac{n^2\pi^2}{a^2} kt\right)\right] + \sigma m_1 \quad \text{....(36a)}$$

$$F_2 = H_o \frac{64}{3} \frac{a}{\pi^3} \sum_1^\infty \frac{2n^2}{(4n^2-1)^2} \left[1 - exp\left(-\frac{n^2\pi^2}{a^2} kt\right)\right] + \sigma m_1 \quad \text{(37a)}$$

$$F_3 = H_o \frac{2a}{\pi^2} \sum_1^\infty \frac{1}{n^2} \left[1 - exp\left(-\frac{n^2\pi^2}{a^2} kt\right)\right] + \sigma m_1 \quad \text{(38a)}$$

$$F_4 = \frac{H_o 2a}{\pi^3} \sum_1^\infty \frac{1}{n^3} \left\{ sin\, n\pi \frac{h}{a} + n\pi (1 - h/a) \cos n\pi \frac{h}{a} \right\}$$
$$\left[1 - exp\left(-\frac{n^2\pi^2}{a^2} kt\right)\right] + \sigma m_1 \quad \text{(39a)}$$

Here the term σm_1 which represents the present surface flux due to the original heat is small compared to the term arising from the radioactivity. It is estimated that σm_1 is between .2 and .4 $\times 10^{-6}$ cal./cm.^{2}sec., depending upon the assumptions as to the conductivity and the initial temperatures. A value of .3 will be adopted as a satisfactory approximation for present purposes. The first quantities in the sums (*i.e.*, those which do not contain the exponential factor) represent the steady-state flux. This is best calculated directly from formulae (29), (31), (33) and (35b), respectively, for steady-state temperatures. The sums containing the exponential factors are rapidly convergent and were separately computed to 20 or more terms, using a value $.01n^2$ for the exponent, which corresponds to $k = .0065$, $t = 2$ billion years; likewise to $k = .0086$, $t = 1\frac{1}{2}$ billion years. The results of these computations for distributions $H(r)$ of type 1′, 2′, and 3′ in Figure 3 are, respectively,

$$F_1 - \sigma m_1 = 9.7 \times 10^6 H_o \text{ (for } n = 1) \quad \text{(48)}$$
$$F_2 - \sigma m_1 = 9.97 \times 10^6 H_o = 2.35 \times 10^7 H_s \quad \text{(49)}$$
$$F_3 - \sigma m_1 = 2.21 \times 10^7 H_s \quad \text{(50)}$$

Here H_s represents the value of the heat generation at the surface, H_o that at the center of the earth. Formulae (49) and (50) are nearly alike. For values of the surface heat, H_s, of order 10^{-13}, they yield somewhat higher values than are now observed for the surface heat flow and indicate that the radioactivity must diminish, rather than remain constant or increase, with depth. However, the uncertainties in present knowledge of the surface heat flux are such that large amounts of radio-activity may exist at great depth without noticeable effect upon the surface heat flow. For example, if in Equation (48) H_o is 10^{-13} cal./cc.sec., as in Deccan or Columbia River basalts, the present flux due to this distribution is only 9.7 $\times 10^{-8}$—namely only about 8 per cent of that being observed. An additional flux of this magnitude would, of course, escape notice, since it is within the present observational error in such

measurements. If this heat were present, the rise in central temperature during the earth's lifetime would be (in accordance with Equation 51), with $\sigma = .01$,

$$T_2(o, t) = \frac{H_o}{\sigma}\frac{a^2}{\pi^2}(.015) = 6{,}200°\text{C} \quad \ldots\ldots\ldots\ldots\ldots\ldots (51)$$

The hypothesis of so great a temperature rise does not seem acceptable.

TABLE 6.—*Surface heat flow*

Produced by a Uniform Radioactive Layer of Thickness h km.
Heat Generation = H cal./cc. sec.
Diffusivity = .0065
Elapsed Time = 2×10^9 years
Initial Temperature, zero.

$\frac{\pi h}{a}$	h km.	Surface heat flow in H × 10^7 cal./cm.^{2} sec.	
		Present	Steady-State
0	0	0	0
.02	40.5	.376	.403
.05	101.4	.862	.998
.10	202.8	1.59	1.96
.20	405.5	2.03	3.80
.50	1014	2.14	8.61
π	6370	2.22	21.2

PRESENT SURFACE HEAT FLOW DUE TO LAYERED DISTRIBUTIONS

The present surface heat flow due to superficial layers of thickness h in which the heat generation rate is H may be obtained by differencing Equations (50) and (39a). In Table 6 are listed the present flows, and also, for comparison, the ultimate flows arising from layers of a number of different assigned thicknesses h. The results are also shown graphically in Figure 4. The significant conclusion from these calculations is that the present flux diverges rapidly from the steady-state value when the layer thickness exceeds 200 km. When the thickness is 400 km., the present flux is about ½ the ultimate value, and further increase in the layer thickness produces almost no increment in the surface flux. By the aid of Table 6 or the curve in Figure 4, the present surface heat flow in earth models having heat generation distributed in uniform layers may be quickly computed. For example, consider the following model, in which the successive layers correspond respectively as to radioactive heat

with (1) granite, (2) Deccan and Columbia River basalt (3) stony meteorites, and (4) iron meteorites.

Layer (km.)	Heat generation in 10^{-13} cal/cc.sec.
0–10	2
10–40	1
40–2900	.5
2900–6370	.25

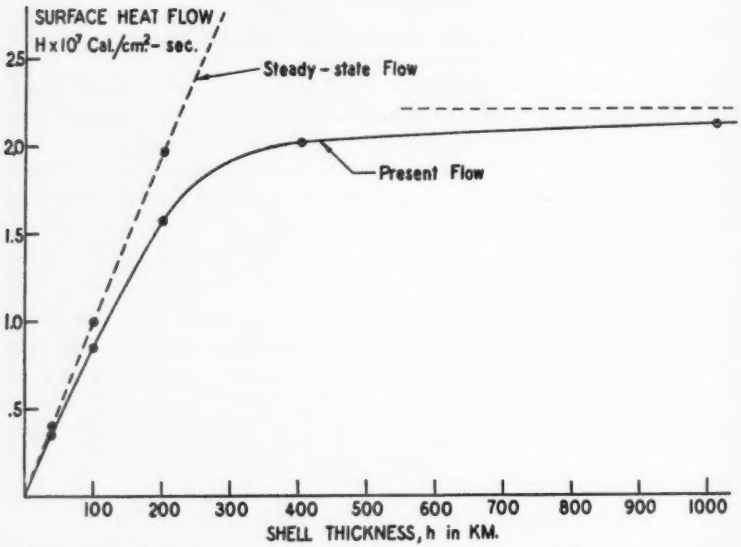

FIGURE 4.—*Surface heat flow produced by a uniform radioactive layer of thickness h in km.*

Heat generation = H cal/cc. sec.; diffusivity = .0065; elapsed time = 2 x 10^9 years; initial temperature, zero.

The model closely resembles, in respect to heat generation, the earth proposed by Washington (1925; 1929). The present surface heat flow produced by this model is 1.38×10^{-6} cal./cm.^{2}sec. If $.3 \times 10^{-6}$ be added to take account of the flux due to the earth's original heat, the total present flux is 1.68×10^{-6}. To see the effect of the depth to which the radioactivity extends, let it be successively assumed that the heat generation in the above model is zero at all depths below 400, 200, and 100 km. respectively. Then the total present surface flows (including in each case an amount $.3 \times 10^{-6}$ due to original heat) for the several cases are as shown in Table 7.

Both Tables 6 and 7, and especially the curve in Figure 4, are of use in furnishing quantitative illustration of several features:

(1) An impracticably high degree of accuracy is needed to distinguish between the effects of different distributions of heat sources at depths below 200 km. An appreciation of the difficulty of estimating the amount of heat generation at depth in the presence of shallow heat sources is

TABLE 7.—*Effect of downward extent of heat generation upon present surface flux*

Example No.	1	2	3	4
Downward extent of heat generation	To Center	400 km.	200 km.	100 km.
Present surface flux, in 10^{-6} cal./cm.^{2}sec.	1.68	1.58	1.37	1.00

furnished by the following example. If the heat generation in the model be assumed uniform throughout, then the zone within 400 km. of the surface will account for 90 per cent of the present heat flow of radioactive origin. The remainder of the earth, comprising 82 per cent of the volume, will contribute only 10 per cent of the flux. In fact, it is clear that the present heat flows produced by deep and shallow heat sources uniformly distributed throughout thin spherical shells are in the ratio of the slopes of the curve in Figure 4 at depth and at the surface. At depths greater than 200 km. the slope diminishes to a very small value. Indeed, at 400 km. the slope is only 4 or 5 per cent of its initial value, which means that the present surface heat flow due to radioactivity at depth 400 km. is only 4 or 5 per cent of that occasioned by an equal, shallow concentration.

(2) At lesser depths than 100 km. or 200 km. the slope is nearly uniform, being 80 per cent of its surface value at depth 100 km. and 60 per cent at depth 200 km. Throughout this zone, therefore, the heat generation is in fair equilibrium with the heat loss at the surface. It follows that the manner of distribution in depth of the radioactivity above the 100 or 200 km. level is without significant effect upon surface heat flows, and only the total amount present above this level counts. The observed heat flows fix definite upper limits for the total radioactivity which may exist within 100 or 200 km. of the surface. For example, an observed flux of 1.2×10^{-6} cal./cm.^{2}sec. corresponds to an average heat generation in the first 100 km. of depth of 1.4×10^{-13} cal./cc. sec. *or less;* or to an average generation of $.75 \times 10^{-13}$ *or less* above the 200 km. level.

Careful studies of the three pertinent quantities: (1) the surface heat flux, (2) the local thickness and character of the layers composing the crust, and (3) the mean radioactive heat generation associated with the crust (all in the same regions) are necessary in order to ascertain the degree to which known shallow distributions of radioactivity are cor-

related with observed heat losses. After estimation of the shallow generation of heat and allowance for the original heat, the residual flux may be associated in a general nonunique manner with heat generation at greater depths. In no area are satisfactory observations upon all three of the above factors now available. Any of the values for the heat flux in the range 1.00×10^{-6} to 1.68×10^{-6} listed in Table 7 might be in approximate agreement with such partial observations as now exist, and any of the four hypotheses concerning the distribution of radioactivity is therefore tenable.

The rise in internal temperatures accompanying deep-seated distributions of radioactivity is, however, an important feature and appears to set certain vaguely understood limits upon the rate of heat generation that may exist at depth. In the model, in which the radioactivity assigned to the core is that in iron meteorites, the temperature rise in the core during 2 billion years is (if $\rho c = 4.0$) about 400° C. In the mantle, the rise would be greater, possibly about 1500° C. If the suggested mode of origin of the mantle be adopted, then the rise permissible without producing remelting is probably measured in hundreds and not thousands of degrees. It then follows that the maximum permissible amounts of heat generation in the mantle at depth are of the order of 10^{-14} cal./cc.sec., or about 10 per cent of that observed in surface plateau basalts. This degree of concentration of the radioactivity toward the surface is by no means implausible and is in accord with geochemical considerations requiring the concentration of the radioactive elements near the surface.

SUMMARY AND CONCLUSIONS

The problem of the cooling of a radioactive earth has been reviewed. It is probable that the mantle solidified from the bottom up, beginning at the boundary of the core. More exhaustive studies of the surface heat flow, of the structure and composition of the crust, and of the mean heat generation in crustal layers, all in the same regions, are desirable. The surface heat flows fix definite upper limits for the amount of radioactivity which may exist within 100 or 200 kilometers of the surface. But thermal observations and theory seem incapable of furnishing information about the thermal state of the earth at depths greater than 200 or 300 kilometers. Little correlation exists between the radioactivity at depth and observed surface heat flux. Heat generation of the order 10^{-14} cal./cc.sec., or 10 per cent of that observed in plateau basalts, may exist at all depths, to the center, without producing effects contrary to the facts of observation. It is unknown whether the earth is heating or cooling at depth. No need is found for a drastic rate of decrease of radioactive substance

with depth. The oft expressed idea that the indicated amounts of radioactivity in the earth are embarrassingly large and greatly exceed the amounts to be expected from observed surface heat flows is fallacious. Hoskins (1839) summarized the thermal problem in these words,

"investigation of the earth's refrigeration . . . still leaves us in a state of perfect uncertainty as to the actual condition of its central parts . . . from want of the experimental determination of values which it must ever be found extremely difficult, if not impossible, to attain with accuracy."

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